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

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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*

\[ \text{SPIN} \uparrow \quad \text{SPIN} \downarrow \]

CDF

SDF
Charge-Density vs. Spin-Density Oscillations

Ga$_{1-x}$Mn$_x$As: Time-resolved Magneto-Optical Kerr Measurements

![Graph showing time-resolved magneto-optical Kerr measurements](image)

- As-grown GaMnAs
- 120-nm Mn: 3% 
- External field: 0.17 T || [100]

D. Wang et al., unpublished
Dependence of the spin wave frequency on field for different directions

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

Angle between the magnetization and applied field
MAGNON SQUEEZING

Spontaneous RS


J. Zhao, A. Bragas, D. Lockwood and RM
COHERENT POLARITONS AND CHERENKOV RADIATION

Propagation Effects
ZnTe: $v = 0.31c$ (subluminal)
LiTaO$_3$ : $v = 3.07c$ (superluminal)

CHERENKOV RADIATION
Anomalous First-to-Zero Sound Crossover in La$_{1-x}$Ca$_x$MnO$_3$

Y. Ren et al., unpublished
Propagating and Surface Avoiding Modes

\[ \Delta R/R \]

Time delay [ps]

Frequency [THz]

Wavevector [10^8 m^-1]

\[ \times 10^{-6} \]

\[ \times 10^{-5} \]

\[ \times 10^{-6} \]

\[ \times 10^{-7} \]

\[ \times 10^{-8} \]

\[ \times 10^{-9} \]

\[ \times 10^{-10} \]
WHAT ARE THE MECHANISMS?
WHAT ARE THE MECHANISMS? DETECTION vs. GENERATION

\[ n \rightarrow n + \Delta n \ (\Delta n \propto Q, Q^2, Q^3, \ldots) \]
LASER-INDUCED IMPULSIVE EXCITATION OF ATOMS

DISPLACEMENTS
(u \equiv \text{ions} ; \ Q \equiv \text{phonons})

CHANGE IN DIELECTRIC RESPONSE

CHANGE IN ENERGY DENSITY

FORCE DENSITY

ELECTROMAGNETIC ENERGY DENSITY

\[ U = \varepsilon \left| E(r,t) \right|^2 / 8\pi \]

\[ \delta \varepsilon = \sum_{im} \left( \partial \varepsilon / \partial u_{im} \right) u_{im} + \sum_{ijmn} \left( \partial^2 \varepsilon / \partial u_{im} \partial u_{jn} \right) u_{im} u_{jn} + \ldots \]

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

\[ \sum_s \left( \partial \varepsilon / \partial Q_{s,k=0} \right) Q_{s,k=0} + \sum_{st,k} \left( \partial^2 \varepsilon / \partial Q_{s,k} \partial Q_{t,-k} \right) Q_{s,k} Q_{t,-k} + \ldots \]

\[ F \propto \left| E^2(t) \right| \]

FIRST-ORDER IMPULSIVE FORCE

SECOND-ORDER IMPULSIVE CHANGE OF FREQUENCY

IMPULSIVE STIMULATED RAMAN SCATTERING
ABOVE THE GAP: DISPLACITIVE EXCITATION

\[ H = \sum_b \varepsilon_b c_b^+ c_b + \frac{1}{2} \sum_q (p_q^2 + \Omega_q^2 Q_q^2) + \sum_{kk'} \Xi_{kk'} Q_{k-k'} c_k^+ c_{k'} \]

\[ \ddot{Q}_q + \Omega_q^2 \langle Q_q \rangle = -\sum_k \Xi_{k,k-q} \langle c_k^+ c_{k-q} \rangle \]

\[ F \propto \langle c_b^+ c_{b-q} \rangle \]

\[ \tau \gg \Omega^{-1} \]

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

TWO RAMAN TENSORS: T. Stevens, J. Kuhl and RM
\[ \lambda = c_T \]

**COHERENT PHONON FIELD**

\[ K = 0 \]

**SQUEEZED PHONON FIELD**

\[ K_1 + K_2 = 0 \]
DECP vs. COHERENT RAMAN SCATTERING

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

\[ F_{RS} \propto \sum_b \langle c_{b-q}^+ c_{b} \rangle \]

incoherent → coherent
A-type modes

Long-lived Raman coherence
All Raman-allowed modes
Good (?)
ERL Idea
Time Resolved EXAFS: Impurity Mode Identification

Silicon K-shell: 1.84 keV

Gallium K-shell: 10.4 keV

**Figure 2.** Polarized micro-Raman spectra in the (x-y) scattering configuration for undoped, Ga, Al, Sb, and Fe doped ZnO thin films on sapphire. The ZnO:Ga* film was grown in N₂O atmosphere, all other samples were grown in O₂ atmosphere. Triangles denote additional modes.
Time Resolved EXAFS: Impurity Mode Identification

![Graph showing $K^2 \chi(K)$ versus $K(\AA)^{-1}$]