

Generating Coherent Excitations with Laser Pulses



Low Energy Density: Coherent Excitations

- Phonons
- Magnons
- Polaritons

Medium Energy Density: Nonlinear Science

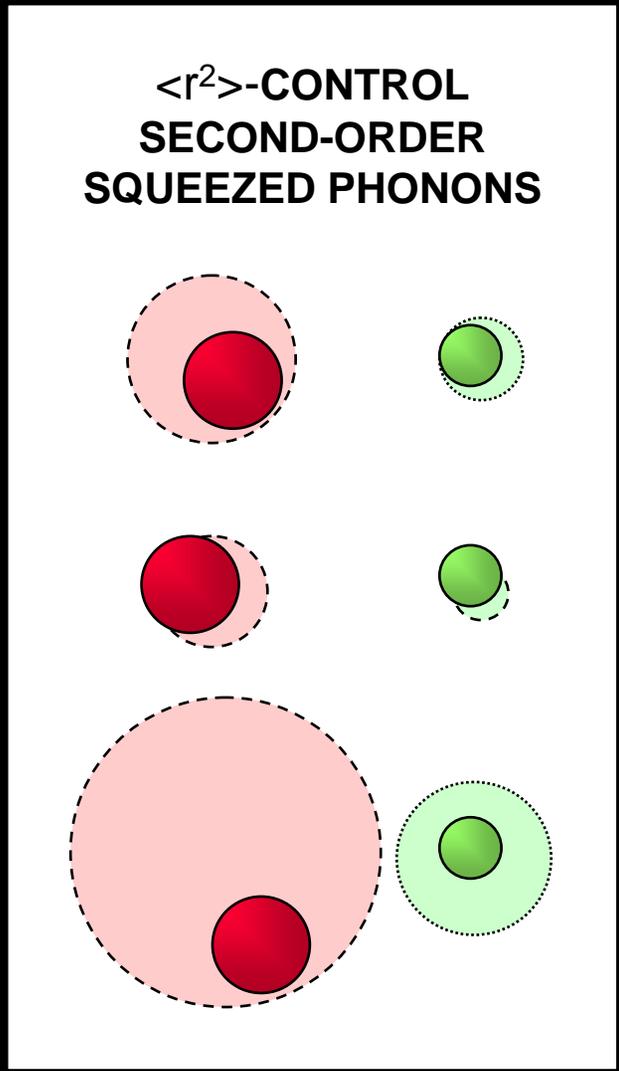
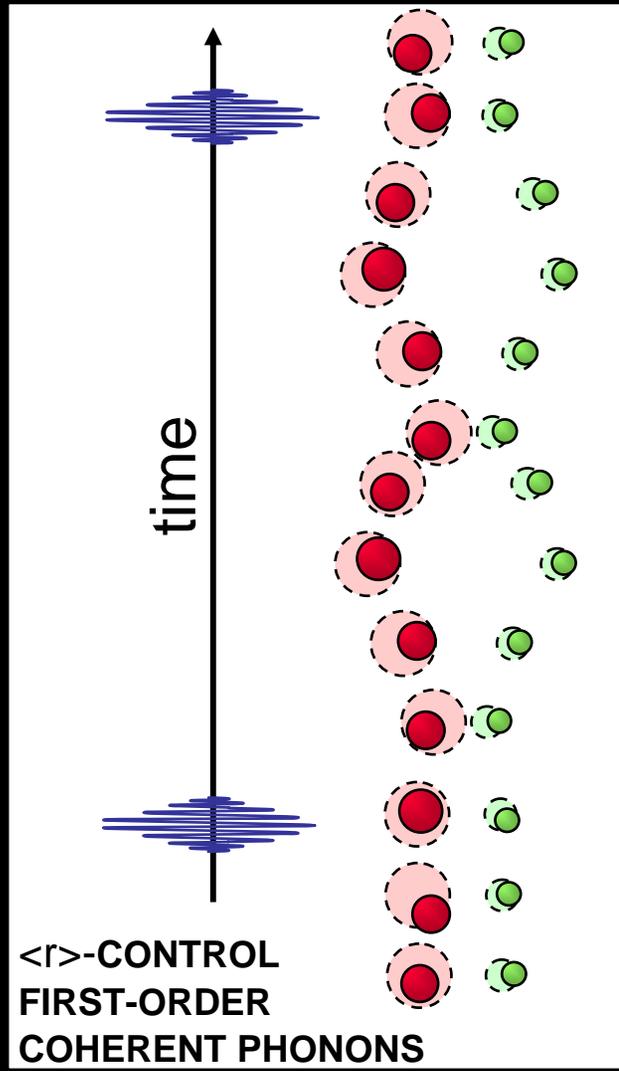
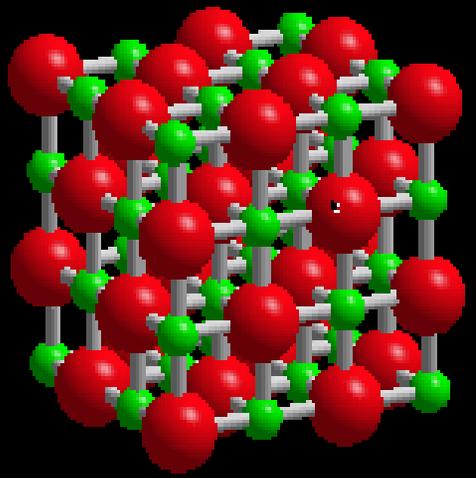
- Anharmonicity
- Non-Thermal Melting
- Dynamical Broken Symmetry
- Domain Reversal

High Energy Density:

- X-Ray Generation
- Laser Ablation
- Shock Compression



PHONONS: NON-PROPAGATING MODES

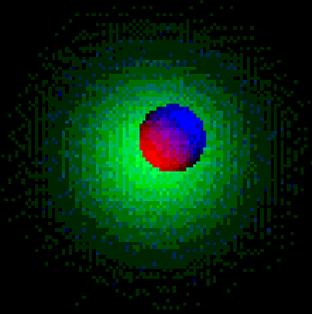


u

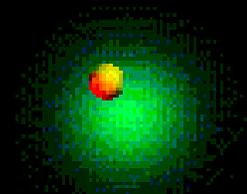
$\langle r \rangle$

$$u^2 = \langle r^2 \rangle - \langle r \rangle^2$$

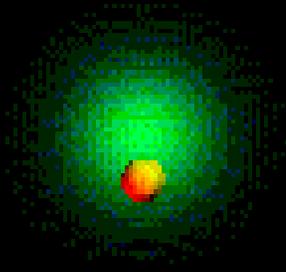
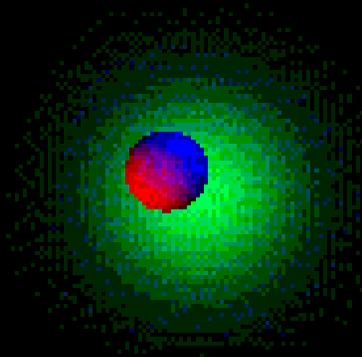
**THERMAL AND QUANTUM
FLUCTUATIONS**



Coherent
Phonon

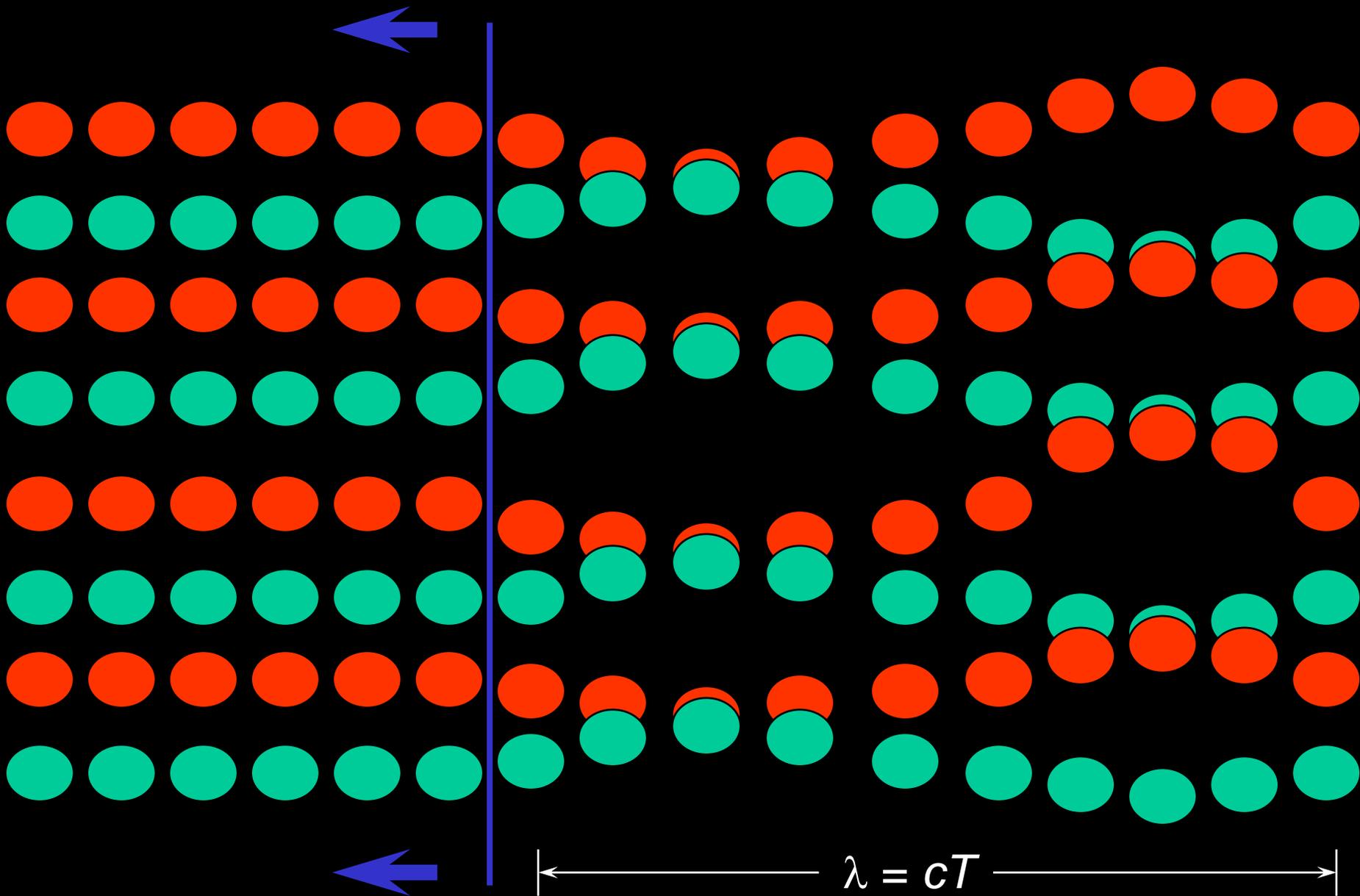


Squeezed
Phonon

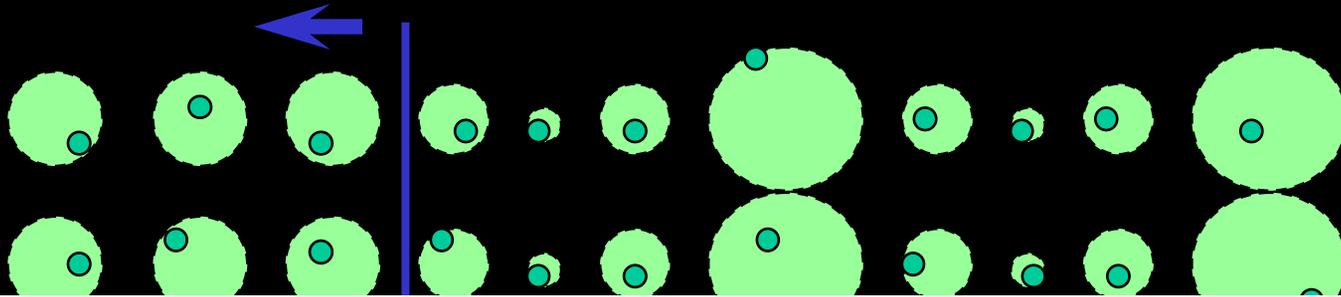


Combined Coherent-Squeezed

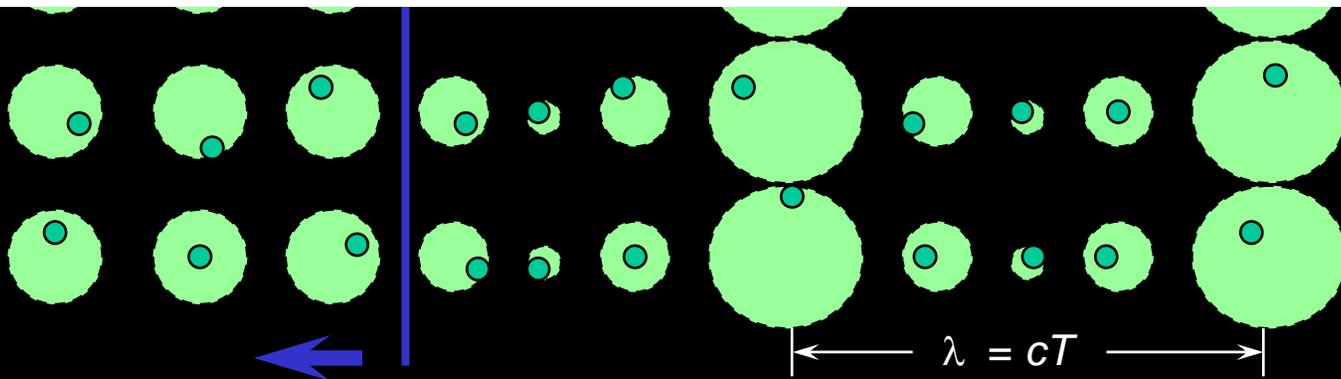
COHERENT PHONON FIELD



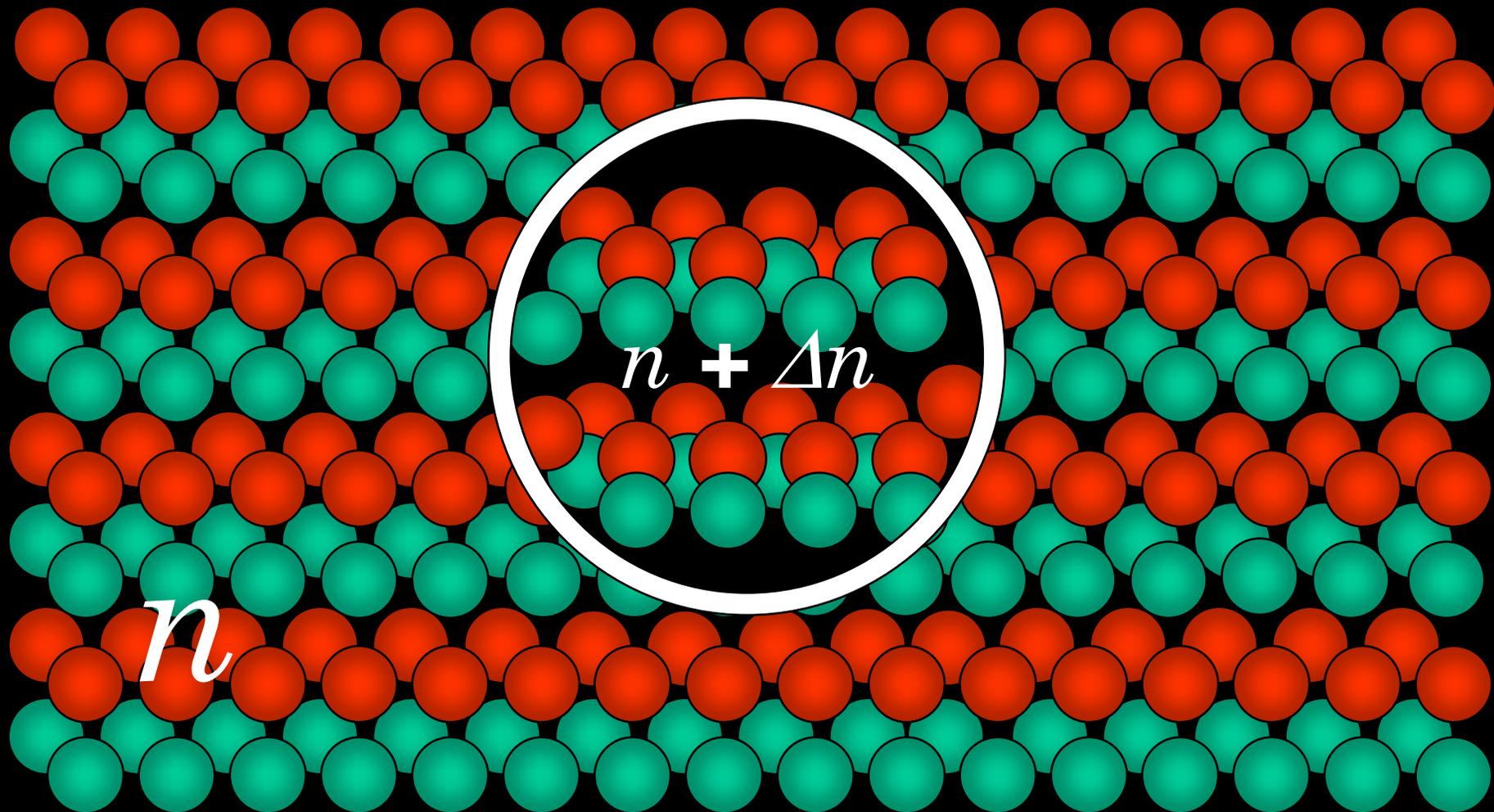
SQUEEZED PHONON FIELD



TIME-VARYING
DEBYE-WALLER FACTOR



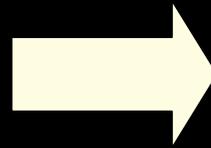
WHAT ARE THE MECHANISMS?



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

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

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

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

FORCE
DENSITY

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

FIRST-ORDER
IMPULSIVE FORCE

$$F \propto Q_{\mathbf{k}} |E^2(t)|$$

SECOND-ORDER
IMPULSIVE CHANGE OF
FREQUENCY

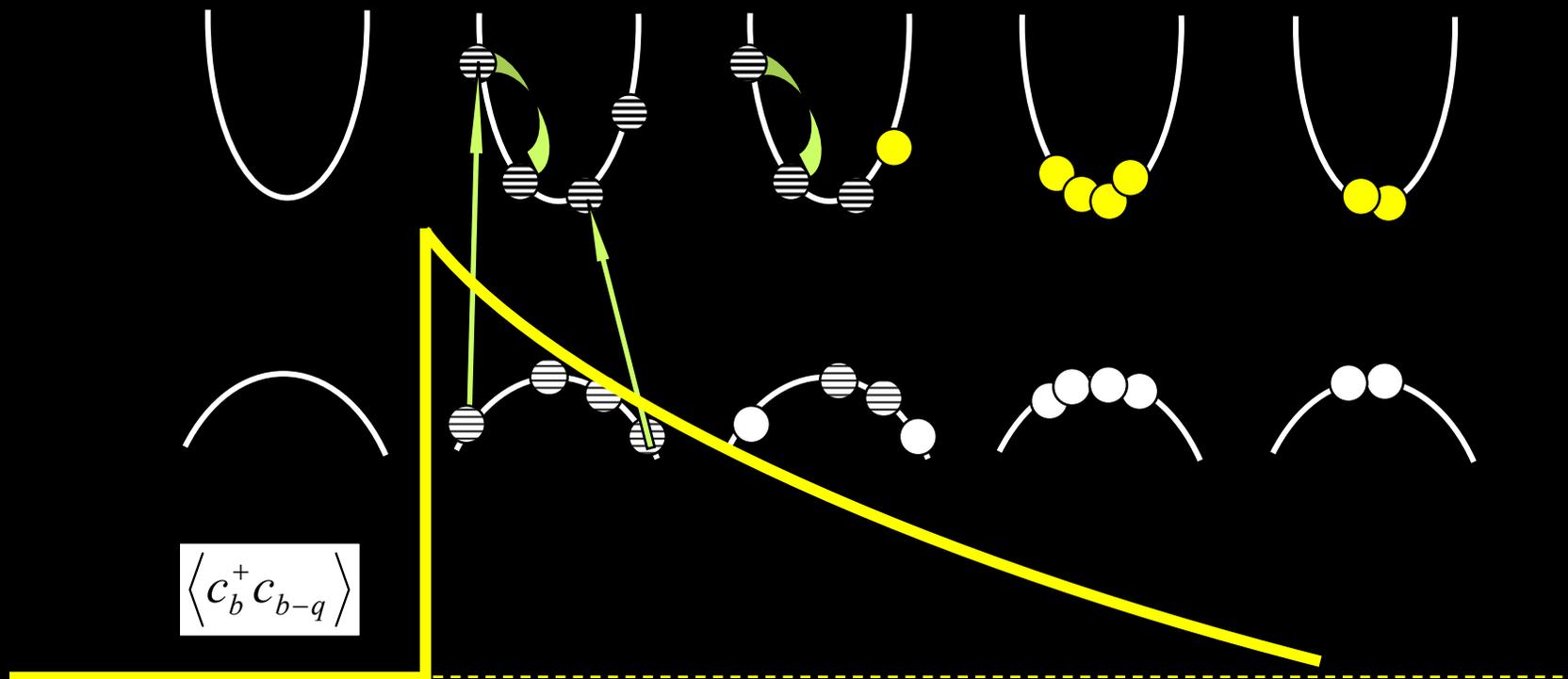
IMPULSIVE STIMULATED RAMAN SCATTERING

ABOVE THE GAP: DISPLACIVE EXCITATION

$$H = \sum_b E_b c_b^\dagger c_b + \frac{1}{2} \sum_q (P_q^2 + \Omega_q^2 Q_q^2) + \sum_{kk'} \Xi_{kk'} Q_{k-k'} c_k^\dagger c_{k'}$$



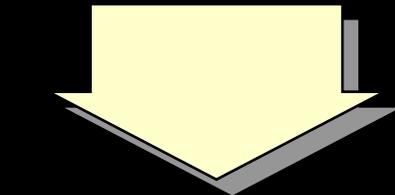
$$\ddot{Q}_q + \Omega_q^2 \langle Q_q \rangle = - \sum_k \Xi_{k, k-q} \langle c_k^\dagger c_{k-q} \rangle$$



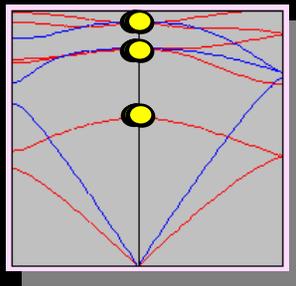
IMAGINARY COMPONENT OF THE RAMAN TENSOR

DISCRETE vs. CONTINUUM

$$\delta U \propto Q_{k \equiv 0} E^2$$



Phonon Energy

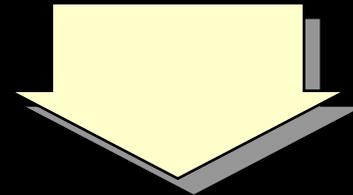


Γ
Wavevector

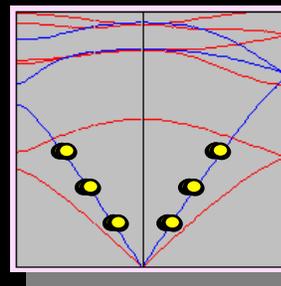
*coherent
phonons*

$$\vec{\mathbf{k}} \approx 0$$

$$\delta U \propto \sum_k Q_k^2 E^2$$



Phonon Energy



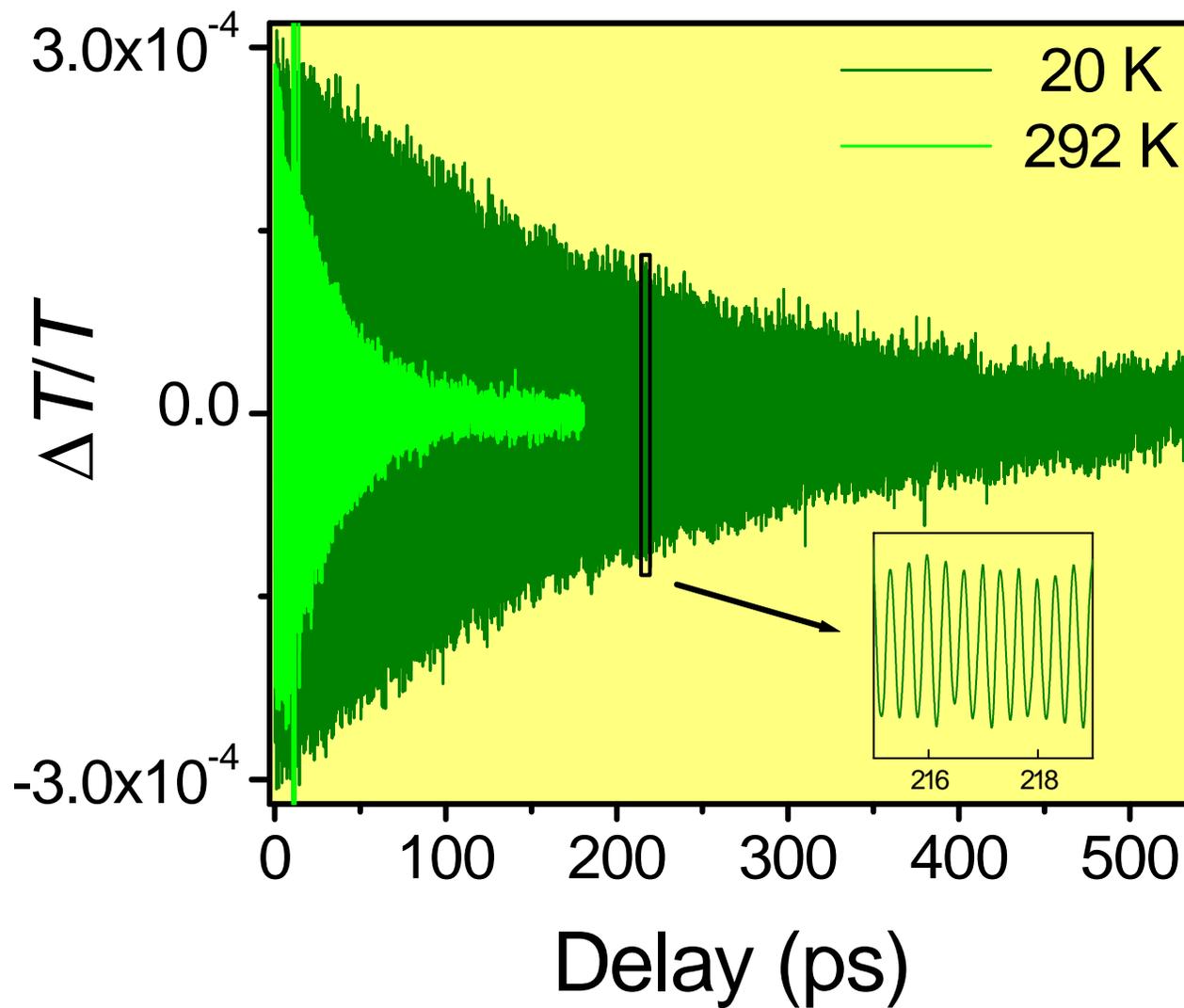
Γ
Wavevector

*squeezed
phonons*

$$\vec{\mathbf{k}}_1 + \vec{\mathbf{k}}_2 \approx 0$$

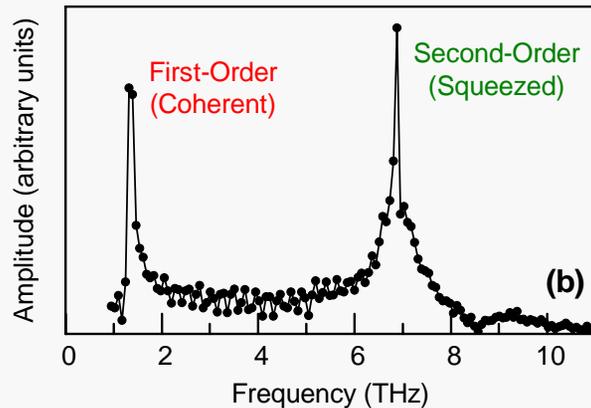
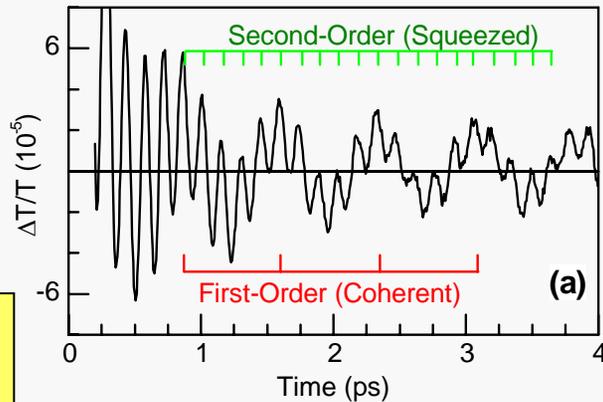
COHERENT PHONONS

ZnO



SQUEEZED AND OTHER CORRELATED FIELDS

SrTiO₃
6K



$$\langle Q_q^{TA} Q_{-q}^{TA} \rangle \neq 0$$

$$\langle Q_q^{TA} Q_{-q}^{TO} \rangle \neq 0$$

KTaO₃

... dispersion compared to the TO₂, therefore zone-boundary mode combinations dominate the ... This is supported by the fact that the difference of

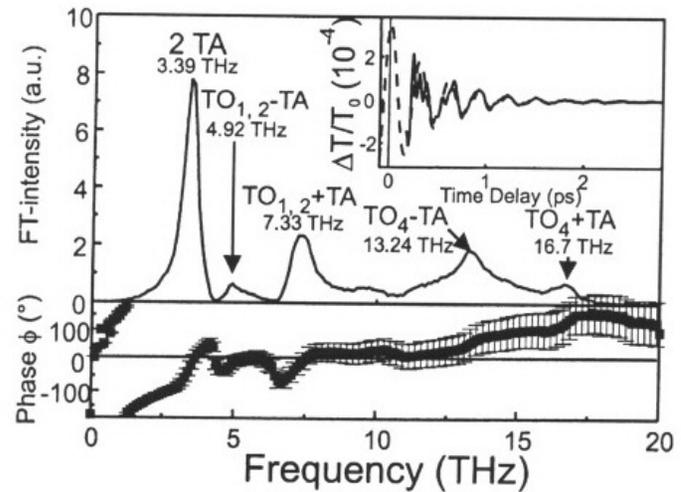
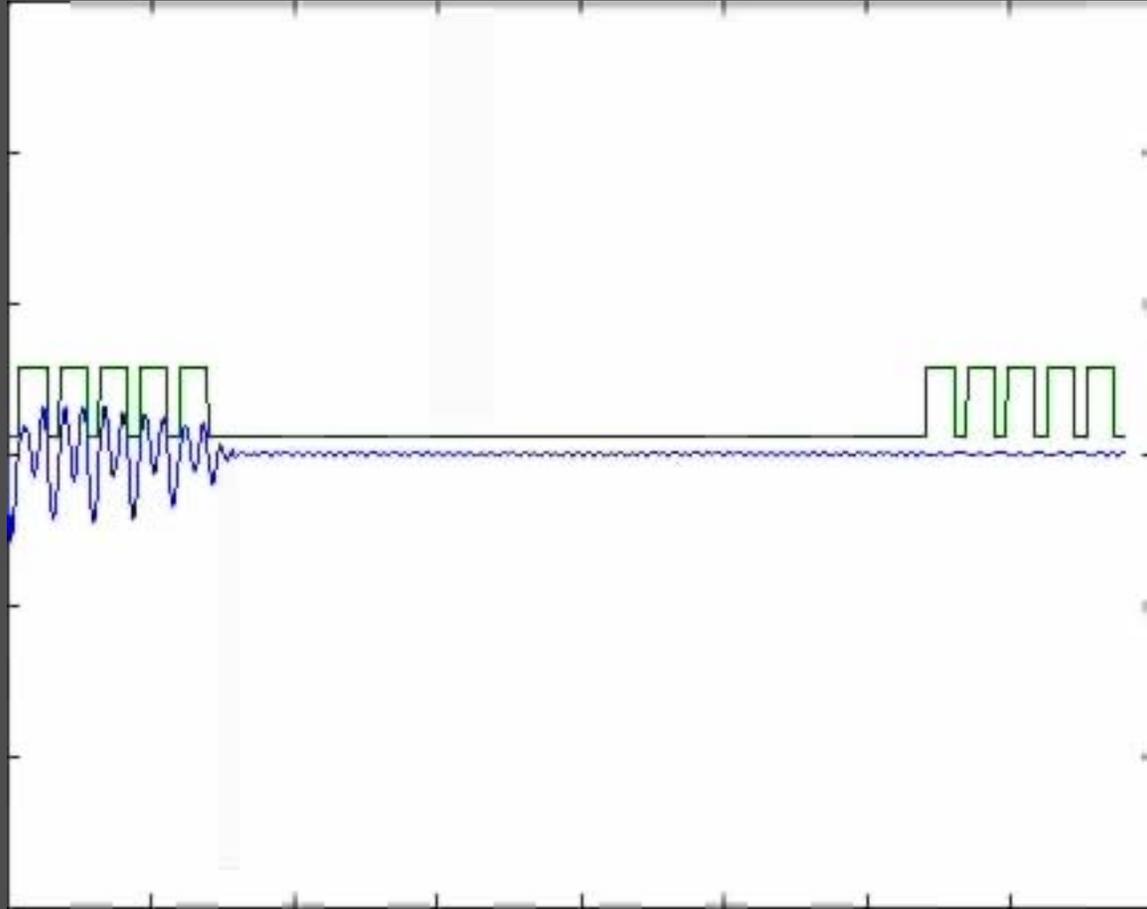


FIG. 1. Fourier intensity and phase spectrum of the transient data at room temperature and a detection wavelength of 835 nm. The excited phonon combinations are indicated in the figure. The phase error is indicated with the error bars. The inset shows the corresponding time-domain data (solid line) and a fit (dashed line) with the 2 TA frequency.

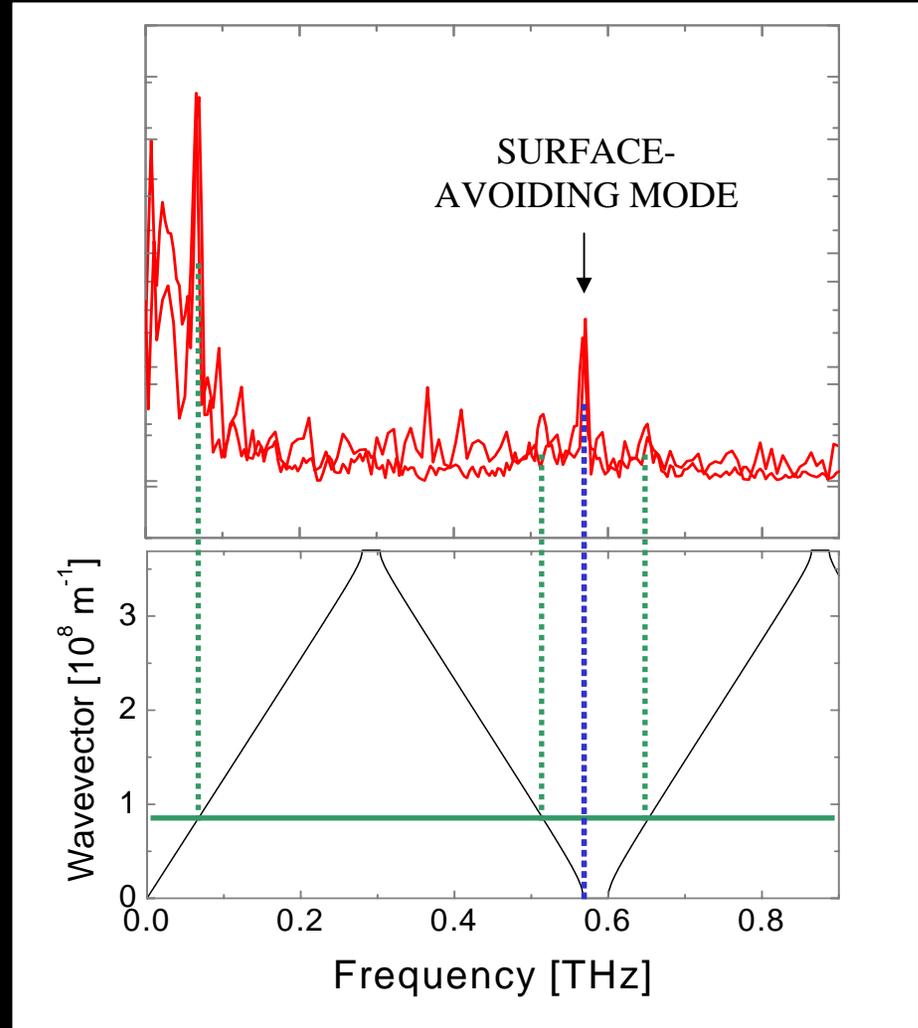
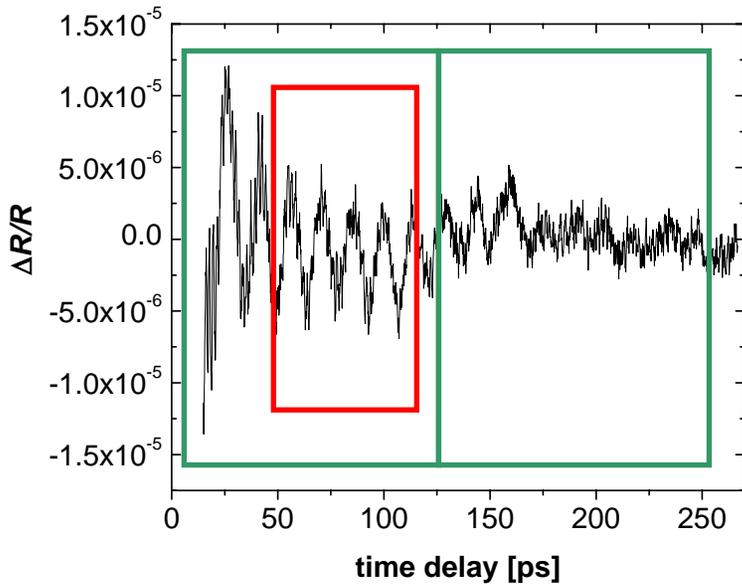
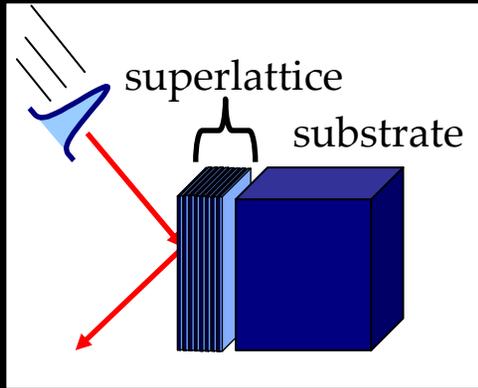
Garrett, Whitaker and Merlin,
in *Ultrafast Phenomena XI*,
(Springer, Berlin, 1998), p.362

Bartels, Dekorsy and Kurz,
Phys. Rev. Lett. **84**, 2981 (2000)

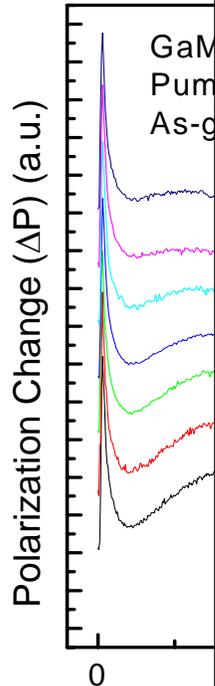
Propagating Acoustic Modes



Propagating and Surface Avoiding Modes



Low-Frequency Magnons: GaMnAs



CONTROL AND USE OF DEFECTS IN MATERIALS **SPECIAL SECTION**

Making Nonmagnetic Semiconductors Ferromagnetic

REVIEW

H. Ohno

Semiconductor devices generally take advantage of the charge of electrons, whereas magnetic materials are used for recording information involving electron spin. To make use of both charge and spin of electrons in semiconductors, a high concentration of magnetic elements can be introduced in nonmagnetic III-V semiconductors currently in use for devices. Low solubility of magnetic elements was overcome by low-temperature nonequilibrium molecular beam epitaxial growth, and ferromagnetic (Ga,Mn)As was realized. Magnetotransport measurements revealed that the magnetic transition temperature can be as high as 110 Kelvin. The origin of the ferromagnetic interaction is discussed. Multilayer heterostructures including resonant tunneling diodes (RTDs) have also successfully been fabricated. The magnetic coupling between two ferromagnetic (Ga,Mn)As films separated by a nonmagnetic layer indicated the critical role of the holes in the magnetic coupling. The magnetic coupling in all semiconductor ferromagnetic/nonmagnetic layered structures, together with the possibility of spin filtering in RTDs, shows the potential of the present material system for exploring new physics and for developing new functionality toward future electronics.

The mass, charge, and spin of electrons in the solid state lay the foundation of the information technology we use today. Integrated circuits and high-frequency devices made of semiconductors, used for information processing and communications, have had great success using the charge of electrons in semiconductors. Mass storage of information—indispensable for information technology—is carried out by magnetic recording (hard disks), magnetic tapes, magneto-optical disks) using spin of electrons in ferromagnetic materials. It is then quite natural to ask if both the charge and spin of electrons can be used to further enhance the performance of devices. We may then be able to use the capability of mass storage and processing of information at the same time. Alternatively, we may be able to inject spin-polarized current into semiconductors to control the spin state of carriers, which may allow us to carry out qubit (quantum bit) operations required for quantum computing (7). However, there are good reasons why this has not yet been realized. The semiconductors used for devices and integrated circuits, such as silicon (Si) and gallium arsenide (GaAs), do not contain magnetic ions and are nonmagnetic (Fig. 1C), and their magnetic g factors are generally rather small. In order for there to be a useful difference in energy between the two possible electron spin orientations, the magnetic fields that would have to be applied are too high for everyday use. Moreover, the crystal structures of magnetic materials are usually quite different from that of the semiconductors used in electronics, which makes both materials incompatible with each other.

Ferromagnetism and semiconductor properties coexist in magnetic semiconductors, such as europium chalcogenides and semiconducting spinels that have a periodic array of magnetic elements (Fig. 1A) (2). In these magnetic semiconductors, which were extensively studied in the late 1960s to early 1970s, exchange interactions between the electrons in the semiconducting band and the localized electrons at the magnetic ions lead to a number of peculiar and interesting properties, such as a small shift of band gap when ferromagnetism sets in. Unfortunately, the crystal structure of such magnetic semiconductors is quite different from that of Si and GaAs; in addition, the crystal growth of these compounds is notoriously difficult. To obtain even a small, single crystal requires weeks of preparation and growth.

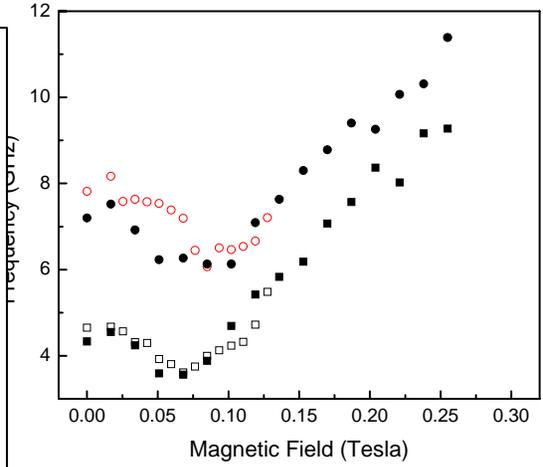
Making Nonmagnetic Semiconductors Magnetic

The usefulness of semiconductors resides in the ability to dope them with impurities to change their properties, usually to p - or n -type. This approach can be followed to introduce magnetic elements into nonmagnetic semiconductors to make them magnetic. This category of semiconductor, called diluted magnetic semiconductors (DMSs; Fig. 1B), are alloys of nonmagnetic semiconductor (Fig. 1C) and magnetic elements (3). Study of DMSs and their heterostructures have centered mostly on II-VI semiconductors, such as CdTe and ZnSe, in which the valence of the cations matches that of the common magnetic ions such as Mn. Although this phenomenon makes these DMSs relatively easy to prepare in bulk form as well as in thin epitaxial layers, II-VI-based DMSs have been difficult to dope to create p - and n -type, which made the material less attractive for applications. The magnetic interaction in II-VI DMSs is dominated by the antiferromagnetic exchange among the Mn spins, which results in the paramagnetic, antiferromagnetic, or spin-glass behavior of the material. It was not possible until very recently to make a II-VI DMS ferromagnetic at low temperature (<2 K) (4).

Ferromagnetic III-V Semiconductors

An approach compatible with the semiconductor used in present-day electronics is to make nonmagnetic III-V semiconductor magnetic, and even ferromagnetic, by introducing a high concentration of magnetic ions. The III-V semiconductors such as GaAs are already in use in a wide variety of electronic equipment in the form of electronic and optoelectronic devices, including cellular phones (microwave transistors), compact disks (semiconductor lasers), and in many other applications. Therefore, the introduction of magnetic III-V semiconductor opens up the possibility of using a variety of magnetic phenomena not present in conventional nonmagnetic III-V semiconductors in the optical and electrical devices already established.

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$T_C \approx 100$ K (3% Mn)

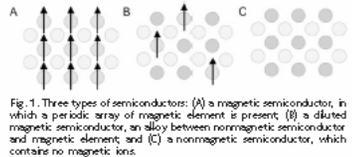
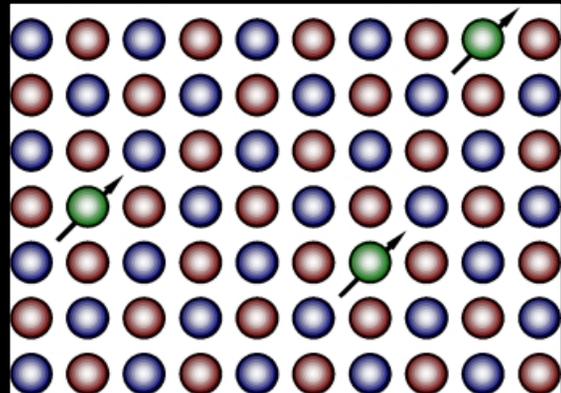
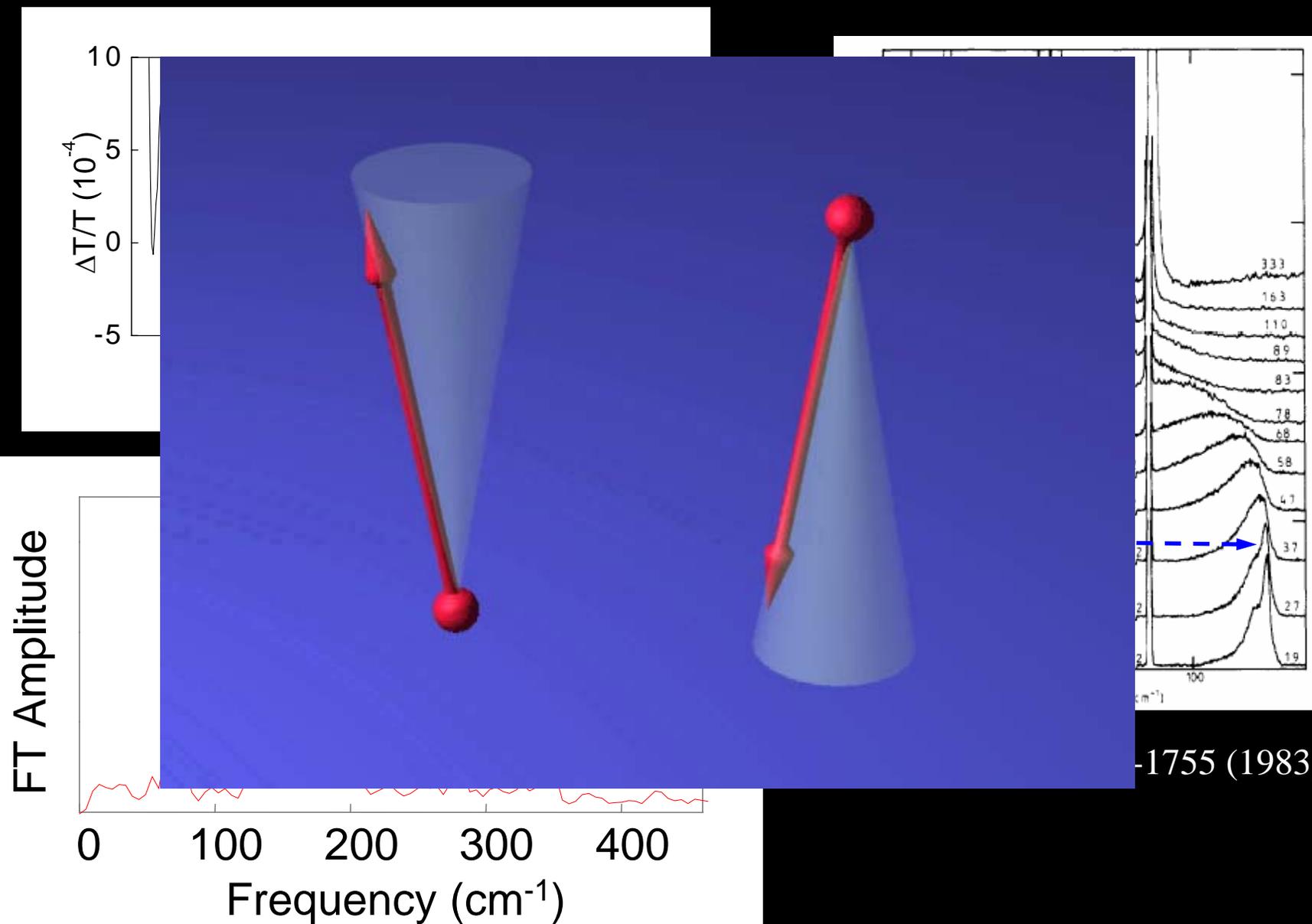


Fig. 1. Three types of semiconductors: (A) a magnetic semiconductor, in which a periodic array of magnetic element is present; (B) a diluted magnetic semiconductor, an alloy between nonmagnetic semiconductor and magnetic element; and (C) a nonmagnetic semiconductor, which contains no magnetic ions.

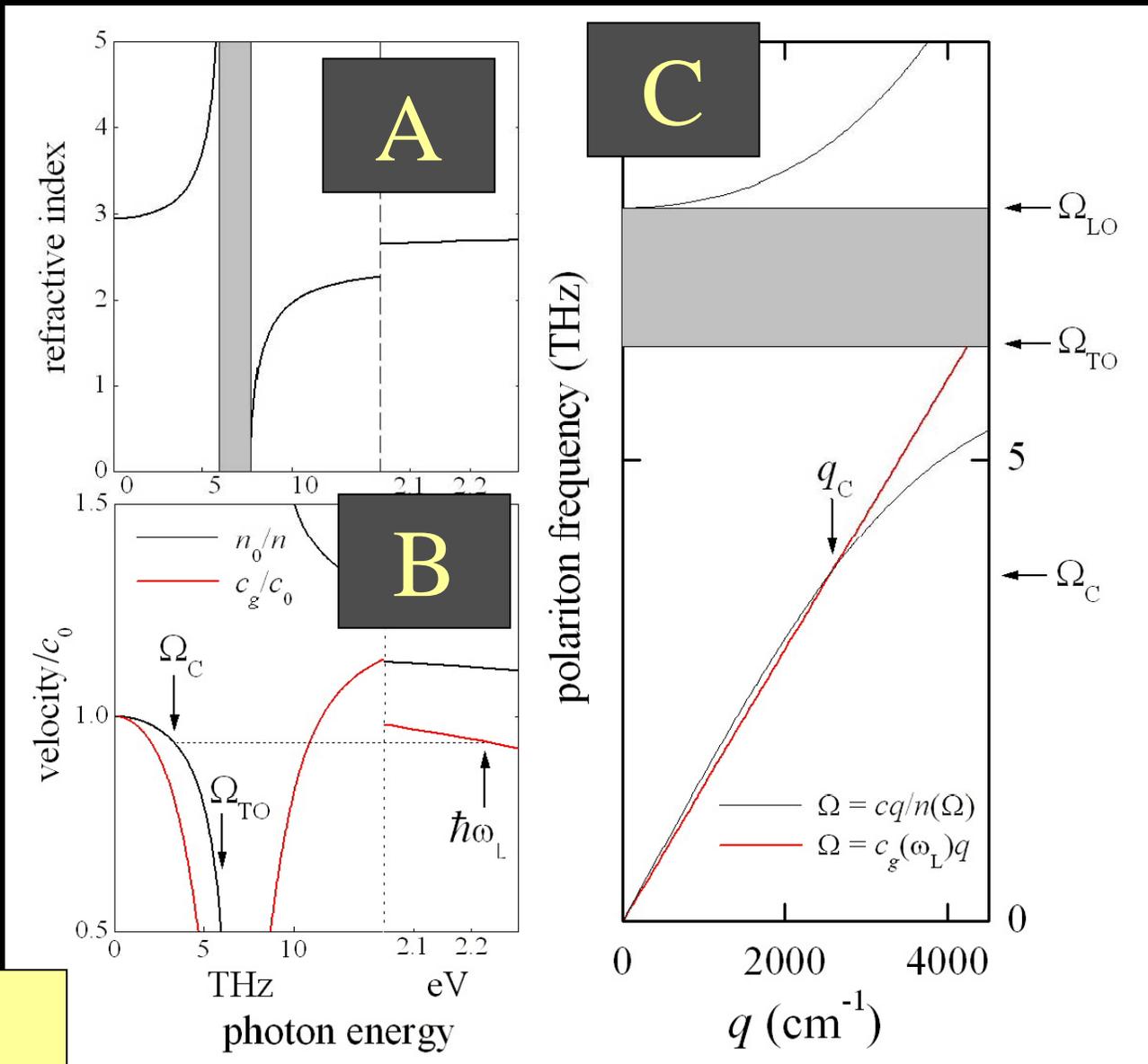


MAGNON SQUEEZING

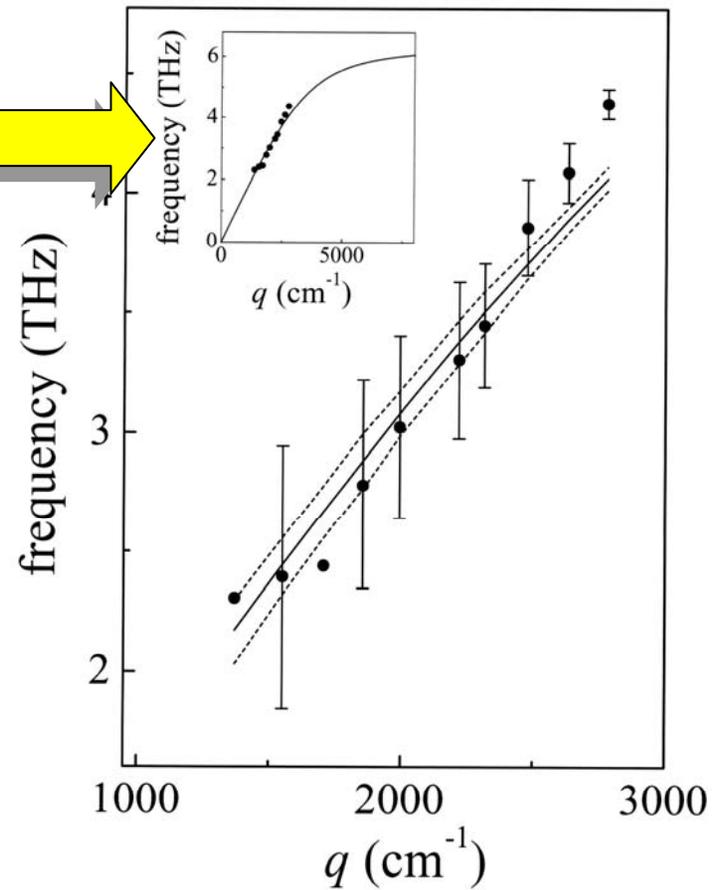
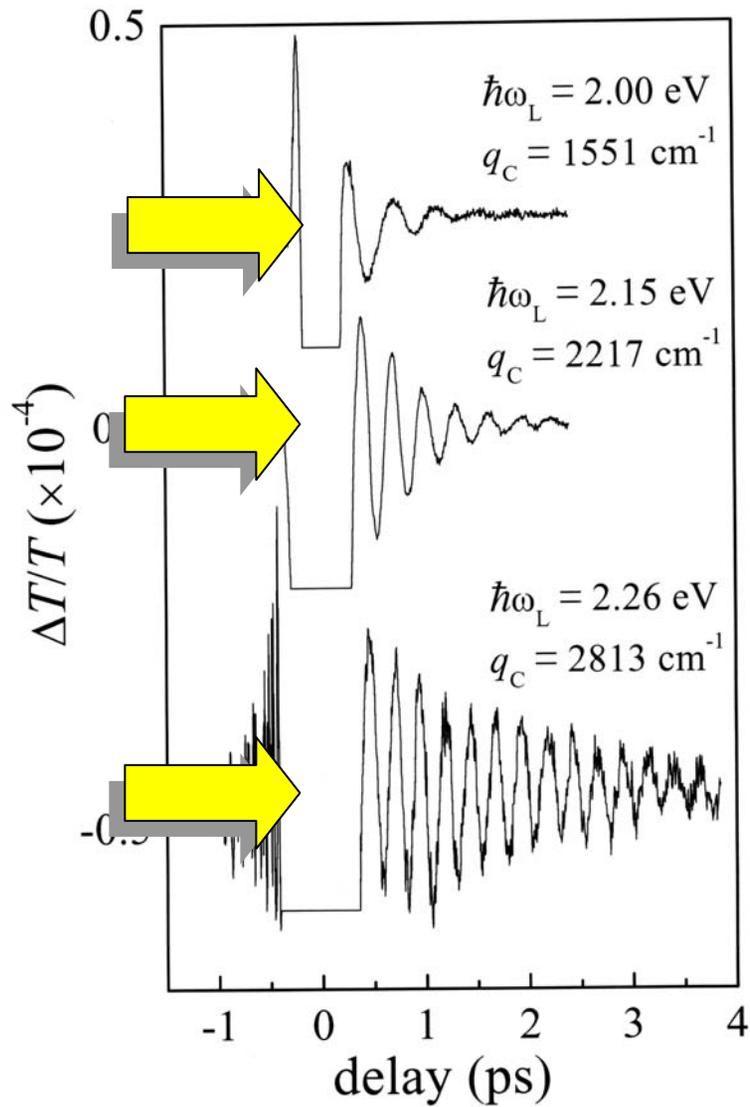
Spontaneous RS



COHERENT POLARITONS

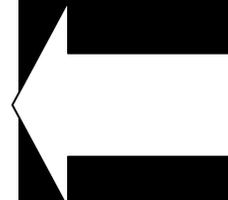
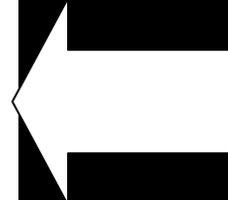
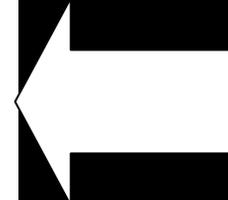
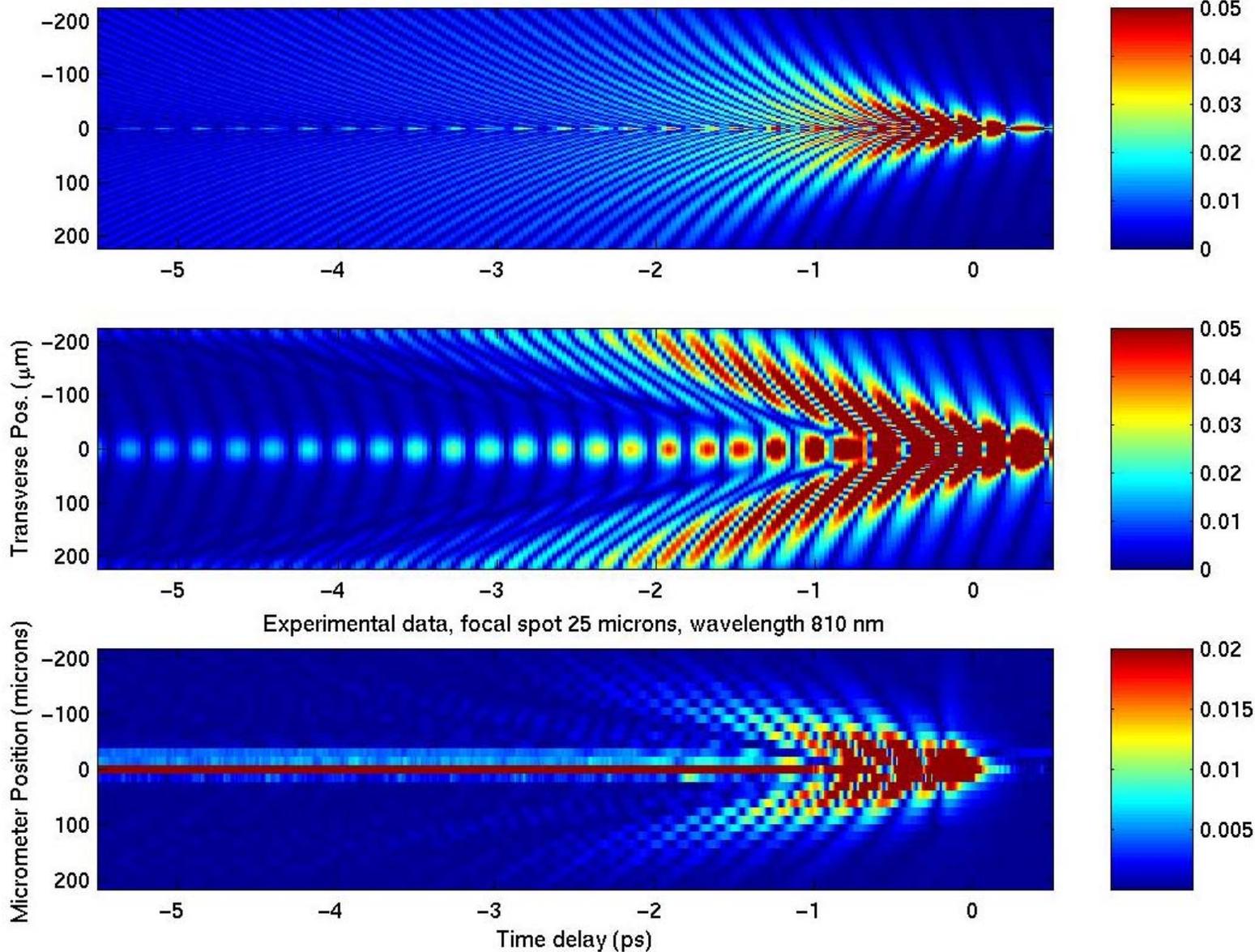


ZnSe

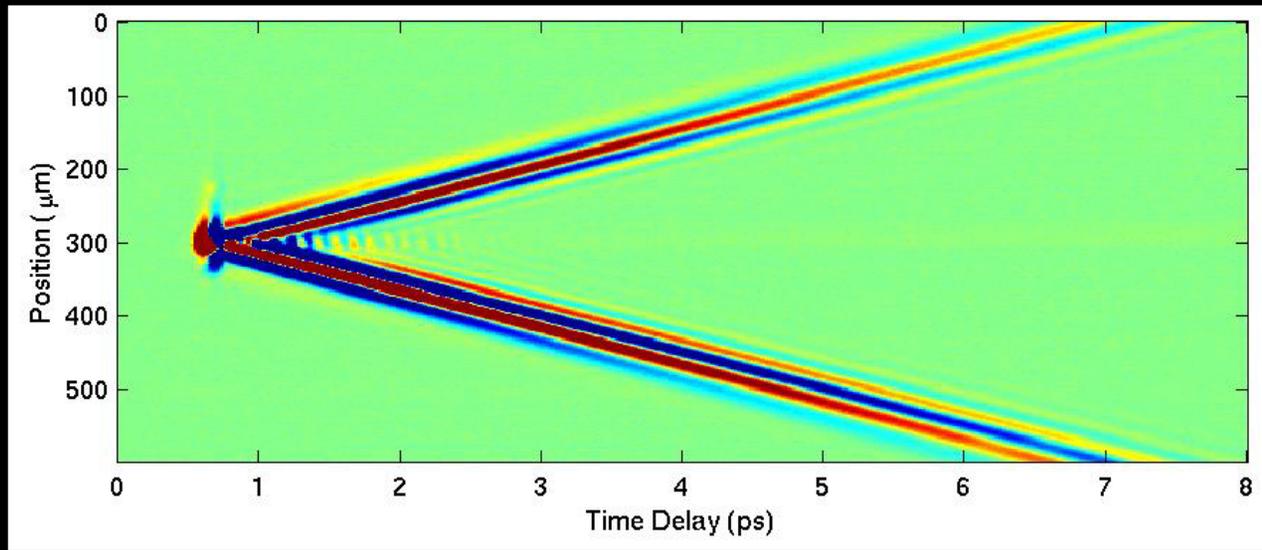


ZnSe

ZnTe: $v = 0.31c$ (subluminal)



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



CHERENKOV
RADIATION

letters to nature

VOLUME 84, NUMBER 1 PHYSICAL REVIEW LETTERS 3 JANUARY 2000

Time-Resolved X-Ray Diffraction from Coherent Phonons during a Laser-Induced Phase Transition

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(Received 10 August 1999)

Time-resolved x-ray diffraction with picosecond temporal resolution is used to observe scattering from impulsively generated coherent acoustic phonons in laser-excited InSb crystals. The observed frequencies and damping rates are in agreement with a model based on dynamical diffraction theory coupled to analytic solutions for the laser-induced strain profile. The results are consistent with a 12 ps fit electron-acoustic phonon coupling time together with an instantaneous component from the deform potential interaction. Above a critical laser fluence, we show that the first step in the transition disordered state is the excitation of large amplitude, coherent acoustic motion.

PACS numbers: 63.20.Kv, 61.10.-i, 64.70.Dv, 78.47.-p

Recent developments in time-resolved x-ray diffraction, using both synchrotron and laser-plasma based sources, have led to the capability of directly observing structural phase transitions, the motion of complex molecules, and chemical reactions, on picosecond time scales. This has resulted in a number of novel experiments, including the investigation of short-pulse laser irradiation of organic films and ultrafast laser-induced phase transitions in semiconductor crystals [1-3]. In experiments of relevance to biology, real-time studies of photoactivated reactions in molecules such as myoglobin and photosynthetic yellow protein (PYP) have been performed [4,5]. Recent experiments utilizing a laser-produced Cu K α source have demonstrated x-ray diffraction from a coherent acoustic pulse induced by laser heating of CuAs [6]; however, while diffraction from expanded and compressed regions of the crystal were observed, no temporal oscillations in the x-ray intensity at the relevant phonon frequencies were recorded.

In this Letter we report the direct observation of x-ray diffraction from laser-induced coherent acoustic phonons at frequencies up to 0.1 THz. The results are in quantitative agreement with simulations based on dynamical diffraction theory, and consistent with an interpretation based on the excitation of coherent phonon states. Moreover, for sufficiently high laser fluences, we observe a reversible, optically induced phase transition which develops on a time scale equal to one-half of a phonon period. We show that the approach to disorder is through the excitation of large-amplitude, coherent lattice motion.

The experimental setup is similar to previous experiments [2, 7]. A bending magnet beam line at the Advanced Light Source synchrotron produces light in a broad spectrum up to photon energies of ~ 10 keV. A Si(111)

monochromator crystal selects a single 2 θ λ with a spectral bandwidth of 1 m eV, which is then directed onto an InSb crystal at the Bragg angle for the (111) reflection. The penetration depth of the laser and x-ray is asymmetrically cut so that the diffracted crystal at a grazing angle of 3°. We use 150 fs, 1 kHz, 800 nm laser, synchrotron visible electron bunches within the synchrotron laser less than 5 ps. The laser is incident on the crystal and overlapped in both space and time with the x-ray pulse. The time-resolved x-ray diffraction is collected by a CCD camera detector triggered by a GaAs switch [8]. A CCD camera records projected onto a phosphor screen. All averaged for a period of about 1 min to 60,000 shots. The resulting temporal camera is 3 ps; this is monitored via femtosecond pulse split off from the x-ray. The entire time history of the diffracted laser excitation is measured at once, in typical pump-probe geometries.

Coherent phonons are manifested as a lattice in time-resolved x-ray diffraction. Fig. 1(a) shows an unperturbed diffraction pattern at 40 arc sec from the Bragg with one for which the laser is incident during the pulse. At a fluence ~ 20 J/m² threshold of 15 mJ/cm² (defined as that in a permanent decrease in the diffracted few seconds), the laser modifies the diffraction pattern and induces temporal strain

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FIG. 1. Geometrical structure factor $S(\mathbf{Q})$ of InSb as a function of the distance of the two lattice sites for diffraction from (111) and (222) planes. The dashed lines denote the acoustic momentum (or an impulsive shift of the equilibrium Bragg distance) upon transition to an excited electronic state.

letters to nature

derived from the face centered distortion of the cubic unit cell. The strain is displaced from the center along the z axis. The strain is stabilized by a small z displacement of the two lattice sites, which introduces a small phase shift of the two lattice sites.

VOLUME 91, NUMBER 16 PHYSICAL REVIEW LETTERS

Transient Strain Driven by a Dense Electron-Hole Plasma

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(Received 31 December 2002)

We measure transient strain in ultrafast laser-excited Ge by time-resolved x-ray anomalous transmission. The development of the coherent strain pulse is dominated by rapid antipolar diffusion. This pulse extends considerably longer than the laser penetration depth because the plasma initially propagates faster than the acoustic modes. X-ray diffraction simulations are in agreement with the observed dynamics.

DOI: 10.1103/PhysRevLett.91.165502

PACS numbers: 63.20.-n, 42.65.Ry, 61.10.Nd

Subpicosecond laser-induced electron-hole plasmas in semiconductors can produce large amplitude lattice strain and rapid loss of translational order. These effects have been studied extensively in ultrafast linear and nonlinear reflectivity experiments [1-6] and, more recently, in time-resolved x-ray Bragg scattering experiments [7-15]. X-ray diffraction has the advantage that it can provide quantitative structural information.

Ultrafast strain propagation was recently used to control the time-resolved anomalous transmission of x rays [16]. The current work uses this new technique as a bulk sensitive structural probe to study the long-standing problem of propagation and coupling to the lattice of a dense electron-hole plasma following laser excitation.

Many previous x-ray experiments [9,10,15] have been analyzed using the thermoelastic model put forward by Thomson *et al.* [17,18] in which the strain is caused by differential thermal expansion. Deviations from this model are discussed in Thomson *et al.* and have been seen with x-ray diffraction [10,12,13,15,16]. In particular, Cavalleri *et al.* [12,13] studied coherent strain near the melting threshold in Ge utilizing x-ray Bragg diffraction. They concluded that the strain is produced over a region which is thick compared to the optical penetration depth, likely due to ambipolar diffusion. However, their experiment was sensitive only to structural changes in the near surface region due to the short x-ray extinction depth.

In the present experiments, the laser intensity is sufficient to impulsively generate a dense electron-hole plasma at the crystal surface, the dynamics of which are governed by ambipolar diffusion [19] and Auger recombination. The plasma couples to the lattice through the deformation potential. Because the electron-hole plasma diffusion is not limited by the sound speed of the material, the strain ρ_{strain} initially advances faster than the sound speed. The resulting acoustic pulse travels into the crystal bulk at the longitudinal sound speed.

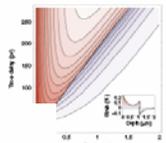


FIG. 1. Geometrical structure factor $S(\mathbf{Q})$ of InSb as a function of the distance of the two lattice sites for diffraction from (111) and (222) planes. The dashed lines denote the acoustic momentum (or an impulsive shift of the equilibrium Bragg distance) upon transition to an excited electronic state.

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(Received 10 August 1999)

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PACS numbers: 63.20.Kv, 61.10.-i, 64.70.Dv, 78.47.-p

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The experimental setup is similar to previous experiments [2, 7]. A bending magnet beam line at the Advanced Light Source synchrotron produces light in a broad spectrum up to photon energies of ~ 10 keV. A Si(111)

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Time-Resolved X-Ray Diffraction from Coherent Phonons during a Laser-Induced Phase Transition

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PH

PHASE TRANSITIONS AND UXD

Femtosecond X-Ray Measurement of Ultrafast Melting and Large Acoustic Transients

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Time-resolved x-ray diffraction with ultrashort (≈ 300 fs), multi-keV x-ray probe study the femtosecond laser-induced solid-to-liquid phase transition in a thin crystal. Nonthermal melting is observed to take place within 300–500 fs. Following observe strong acoustic perturbations evolving on a picosecond time scale.

DOI: 10.1103/PhysRevLett.87.225701

PACS numbers

Femtosecond laser excitation is an ideal tool for driving materials into extremely high nonequilibrium states. Excitation conditions corresponding to one excited electron per atom can be achieved in a time determined by the duration of the laser pulses. Both these states and also the short-lived states of high pressures and temperatures resulting from the thermalization of the initial energy distribution represent exciting areas of study.

One interesting example of this class of phenomena is the electronically induced solid-to-liquid transition in semiconductors, or nonthermal melting [1–3]. This process is triggered by interband excitation of a dense electron-hole plasma ($\approx 10^{22}$ cm $^{-3}$) leading to ultrafast disordering of the semiconductor crystal [4,5] and the formation of a hot liquid at high pressure. A complex chain of phenomena follows on a picosecond time scale, including strong density perturbations propagating away from the molten layer, and the ablation of macroscopic amounts of material [6] at higher laser fluences.

Until recently, ultrafast processes could be measured only by using optical techniques. With the advent of ultrashort x-ray pulses, powerful tools such as time-resolved x-ray diffraction have become available, enabling much deeper investigation into the phenomena of interest here. So far, the power of this new experimental technique has been demonstrated in studies on lattice dynamics [7–10] and ultrafast disordering [11]. Two recent experimental studies have shown that ultrafast melting occurs homogeneously over macroscopic volumes of material [12] and on a subpicosecond time scale [13].

Here we report on a set of experiments that extend the scope of this research both in terms of the investigated phenomena and experimental capabilities. By measuring femtosecond x-ray diffraction from a Ge-Si heterostructure undergoing ultrafast melting at the surface, we directly

measure the disordering recently been reported [12]. Large amplitude transients

VIEW LETTERS

Integrated reflectivity

FIG. 1. versus delay after the femtosecond laser excitation for rapid

Femtosecond Structural Dynamics in VO₂ during an Ultrafast Solid-Solid Phase Transition

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Ultrafast laser pulses were used to probe structural and electronic dynamics during the solid-to-metal transition in VO₂. For high intensity electronic excitation ($\sim 5 \times 10^{21}$ cm $^{-2}$) the material is observed to undergo a solid-to-metal transition. The fast time scale observed suggests that in this case the transition is not thermally initiated.

PACS numbers: 78.47.-g, 71.30.+h, 78.30.Ck

Insulator-to-metal transition in VO₂ (T_M = 340 K) is the latter case, associated with a structural transition from a monoclinic to a tetragonal phase. The fluence of this transition is significantly lower than the measured single-shot damage threshold of ≈ 3 J/cm². Figure 1 shows spatially dependent reflectivity curves, measured along the vertical direction on the individual images. The laser pulses were spatially filtered before interaction with the sample, resulting in a single-transverse-mode, Gaussian spatial profile. Thus, different positions corresponded to different local excitation fluences that were precisely known. As immediately evident in the plot, the reflectivity of the center drops most rapidly toward the equilibrium reflectivity of the metallic phase [6], reached well within hundreds of femtoseconds. At a time delay of about 10 ps, a larger area exhibits the same reflectivity, as parts of the sample that are pumped at lower fluence switch at a slower rate. The low-reflectivity area progressively enlarges, reaching a maximum after approximately 5 ns and slowly returning toward the optical properties of the low-temperature phase with an insulator-to-metal transition time scale of ≈ 10 ns, which is comparable to the time scale of the photoinduced phase transformations [7], above a distinct threshold of the photoinduced phase transition. This, a true phase change can be hypothesized, as opposed to simple carrier excitation, observed at the earliest time delays when the optical properties follow the spatial profile of the pump laser. A 7-nJ/cm² threshold was obtained by comparing the measured maximum transformed area and the pump-fluence profile [8]. Reflectivity evolutions sampled at three different positions of the photopumped spot are shown in the respective insets. Exponential fits to these curves indicate that for increasing pump fluence the insulator-to-metal transition time decreases from more than 50 ps to about 100 fs. A sharp decrease in

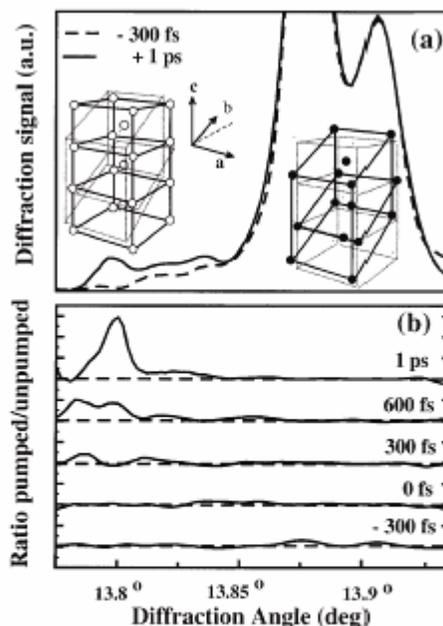


FIG. 2. Upper plot (a): Angle dependent x-ray diffraction signal measured for negative and positive time delays. The diffraction profiles originate from the convolution between the rocking curve of the dynamically strained/transforming crystal and the spectrum of the x-ray plasma source. The dashed and continuous curves correspond to delays of -300 fs and +1 ps, respectively. The two crystalline structures (monoclinic insulating phase and tetragonal metallic phase) are sketched by displaying only the vanadium atoms, which dominate the diffraction signal. Lower plot (b): Ratio between the x-ray reflectivity of the excited and unpumped crystal.

ANHARMONICITY

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Ti₂O₃

LETTERS

11 FEBRUARY 2002

Dynamics of Coherent A_{1g}

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We have investigated the dynamics of the coherent A_{1g} mode under high density excitation. The frequency of the coherent A_{1g} mode decreases as the pump power density is increased, and the frequency shift becomes asymmetric, and the frequency shift with a wavelet transform amplitude of the oscillation, which

DOI: 10.1103/PhysRevLett.88.067401

High density excitation of femtosecond laser pulses has led to the observation of phase transitions, such as femtosecond laser-induced phase transition [1–3]. These phenomena in gap materials are considered to be induced by the excitation of electrons from bonding into antibonding orbitals (electronic softening), which will lead to a distortion along the lattice potential [3,4]. The excitation of electrons changes the equilibrium positions of the atoms; the atoms then oscillate around their new positions, a mechanism known as displacement of coherent phonons [5]. In the past, coherent phonons have been actively studied in various materials from various viewpoints [5–9]. How the dynamics of coherent phonons change under high density excitation with several mJ/cm², the coherent lattice displacement could increase to a significant fraction of the lattice constant. In this case, the amplitude of coherent phonons would play an important role in structural modification. The purpose of this study is to investigate the characteristic properties of the coherent phonons under the anharmonic excitation. This is based on the classical mechanics, which predicts that the frequency of the anharmonic oscillation depends on the amplitude of the oscillation.

The coherent lattice vibrations excited under high density conditions in LiTaO₃, a ferroelectric material transparent to the visible and near-infrared region, were studied by Brennan and Nelson [12]. They observed the overtones of the coherent lattice vibration under high density excitation. However, a frequency shift of the phonon mode due to anharmonicity has not been observed. In this study, we have reported that in photoexcited LiTaO₃, the frequency of the coherent A_{1g} mode decreases as the pump power density is increased. They also reported that the origin of the frequency shift is the anharmonicity of the lattice potential, whereas the asymmetric line shape

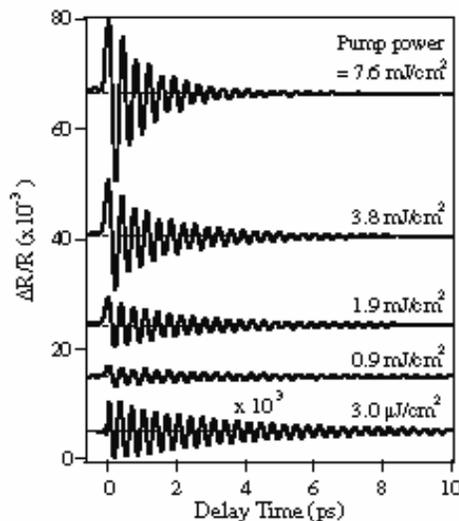
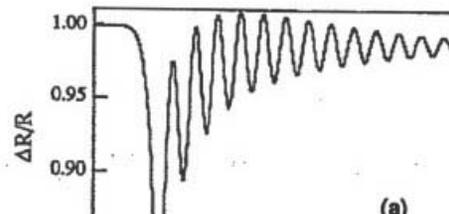


FIG. 1. The transient reflectivity change for Bi at various pump power densities. The dashed lines correspond to the zero level on the reflectivity change of each time-domain signal.

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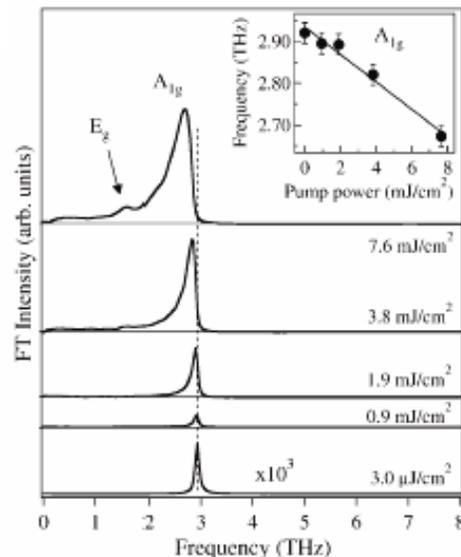


FIG. 2. Fourier transformed spectra obtained from time-domain data in Fig. 1. The dashed line represents the A_{1g} mode frequency of 2.92 THz. Inset: the peak frequency of the A_{1g} mode as a function of the pump power density. The solid line represents a numerical fitting of the data by a linear function.

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SUMMARY

COHERENT EXCITATIONS:

- ALL-OPTICAL PHONON EXPERIMENTS ARE FAIRLY WELL UNDERSTOOD (BUT QUESTIONS REMAIN ABOUT THE GENERATING MECHANISMS: STIMULATED RAMAN vs. OTHERS)
- NEED ULTRAFAST X-RAY DATA TO GET ATOMIC DISPLACEMENTS
- IMPORTANT X-RAY EXPERIMENTS HAVE BEEN PERFORMED BUT MUCH REMAINS TO BE DONE. NOTHING YET ON SPIN EXCITATIONS OR OTHER NON-VIBRATIONAL MODES.

ANHARMONICITY:

- POORLY UNDERSTOOD. PREVIOUS OBSERVATIONS MOST LIKELY DUE TO INDIRECT ELECTRONIC EFFECTS. X-RAY WORK IMPORTANT TO DISTINGUISH VIBRATIONAL FROM OTHER EFFECTS.

NON-THERMAL MELTING:

- VERY IMPORTANT BUT POORLY UNDERSTOOD. X-RAY DATA CRUCIAL. NEED OPERATIONAL DEFINITION. POSSIBLY SERIOUS PROBLEMS WITH TIME SCALES.

TRANSIENT PHASE TRANSITIONS:

- DYNAMICAL BROKEN SYMMETRY
- DOMAIN REVERSAL