THz investigations of exotic quantum states of matter

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U.S. DEPARTMENT OF ENERGY
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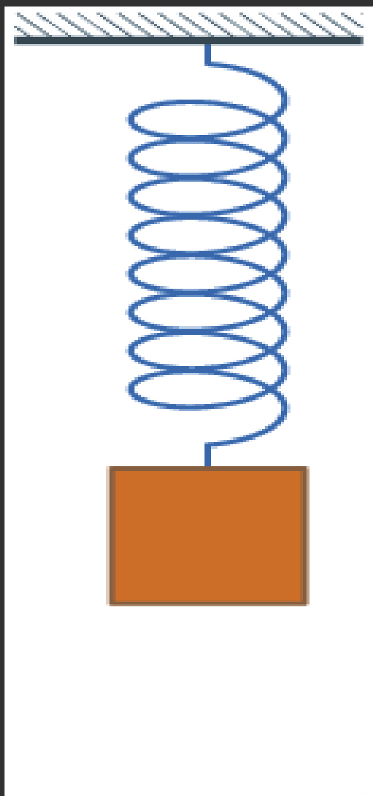
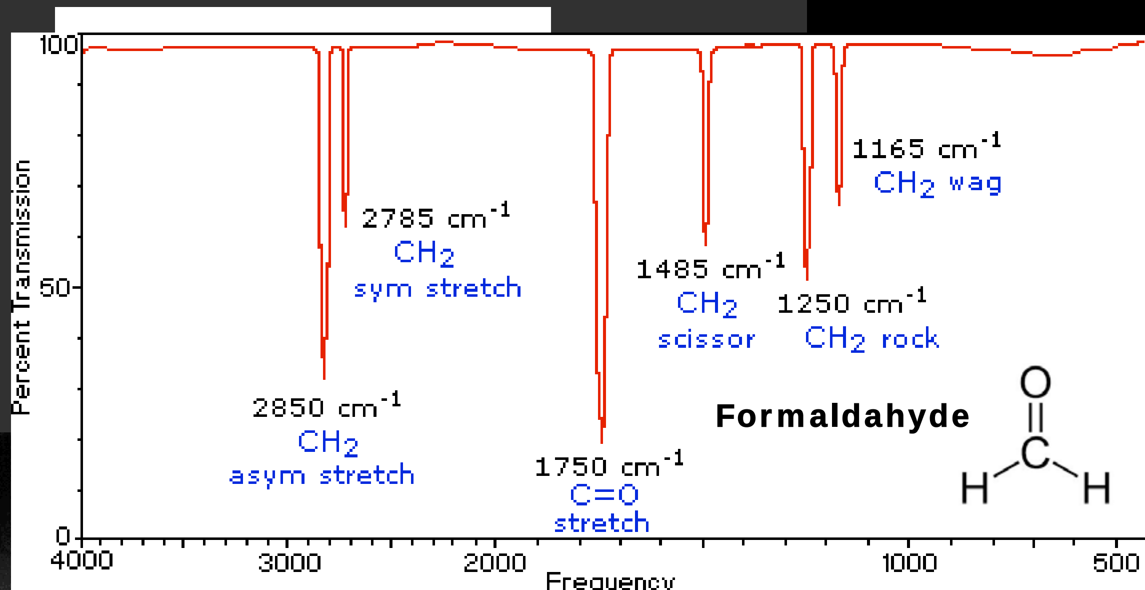
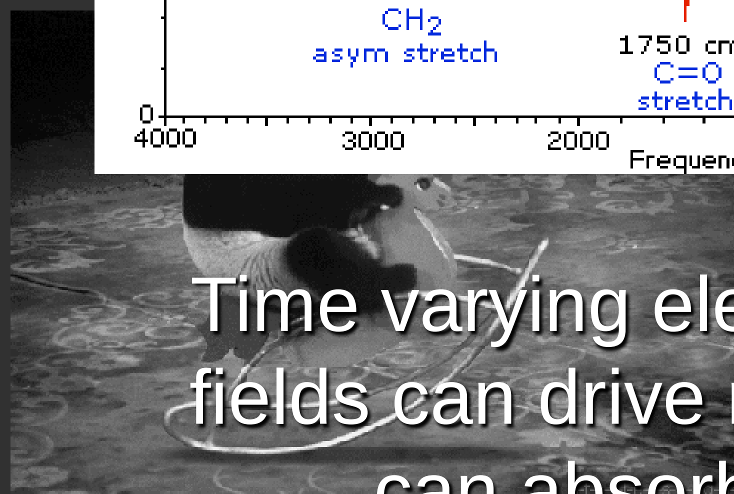
Much of what we know of physical systems comes from their response to finite frequency perturbations…

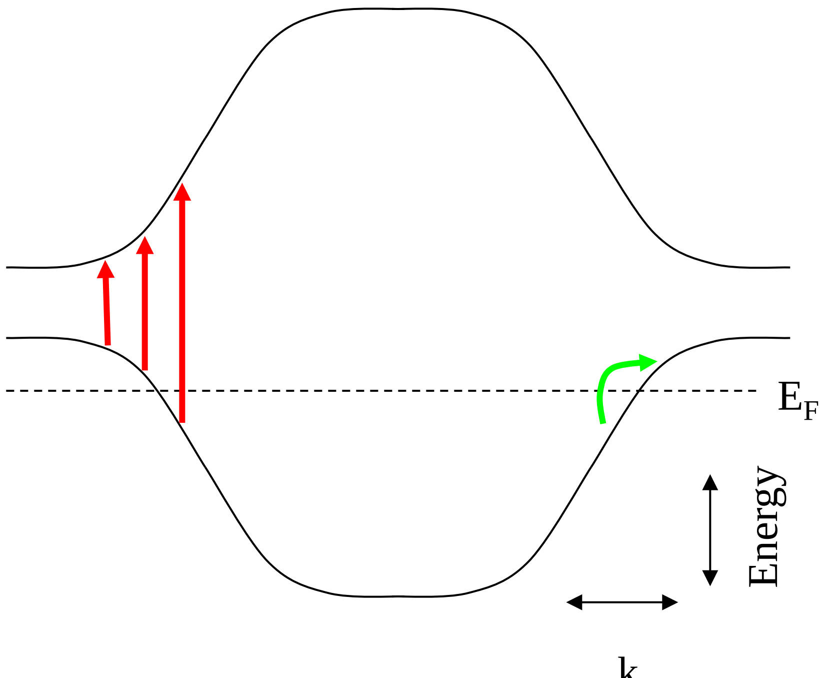


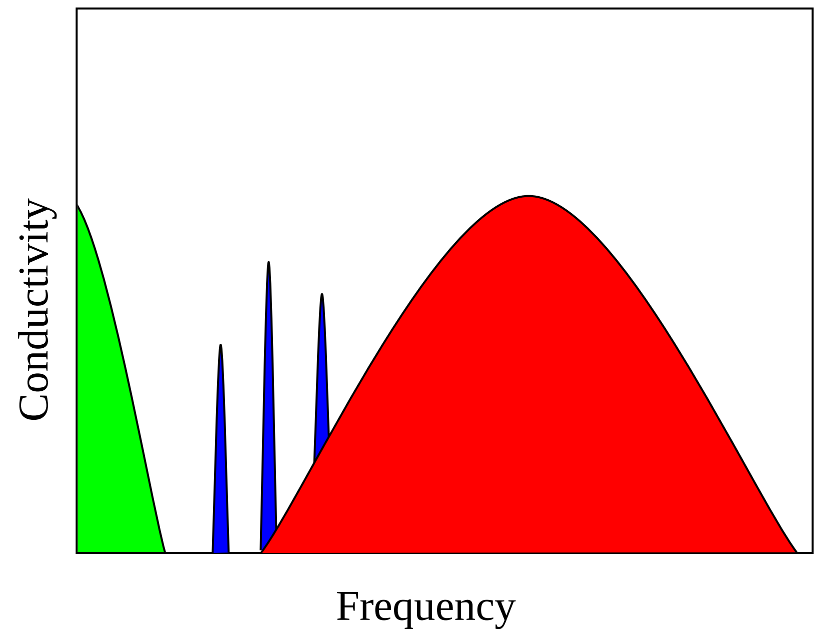




Time varying electromagnetic fields can drive motion, which can absorb energy

Optical Response of Condensed Matter @ q~0





-Intraband Drude response
-Interband response
-Collective Modes (phonons, magnons, excitons etc.)

Time Varying E field (B field) drives electric (magnetic) dipole motion →

$J(\omega) = \sigma(\omega) E(\omega)$

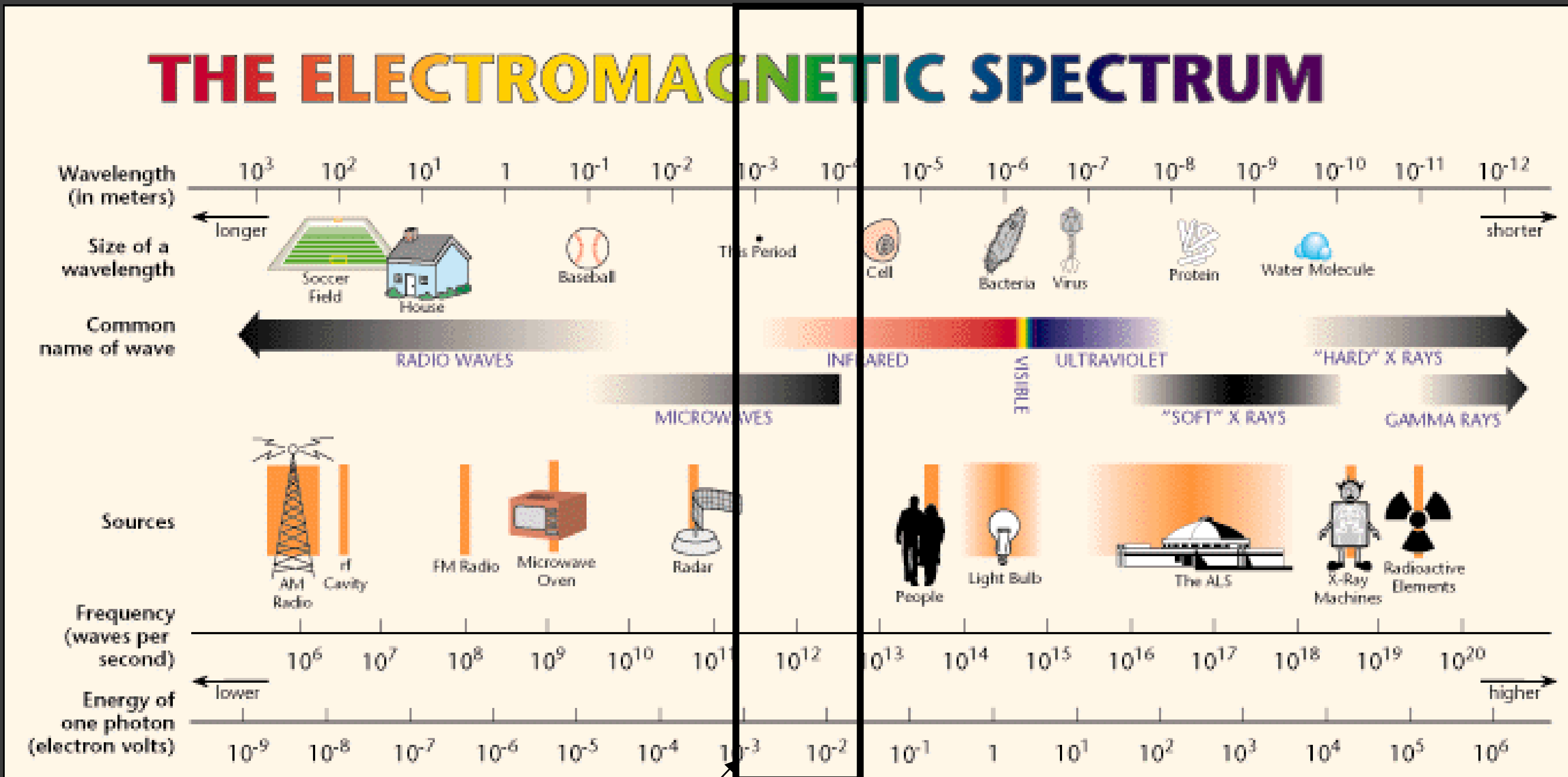
$P = \varepsilon E$ $M = \chi H$

$$\sigma_1^T(\omega) = \frac{\pi e^2}{m^2\omega}\frac{2}{(2\pi)^3}|<s|p|s'>|^2 D_{s's}(\hbar\omega)$$

Application of THz to correlated systems

Electronics

Photonics



10 K

The THz "Gap" 50 GHz – 4 THz

Hard to probe with and detect EM radiation in this range

Q: Why use the THz?

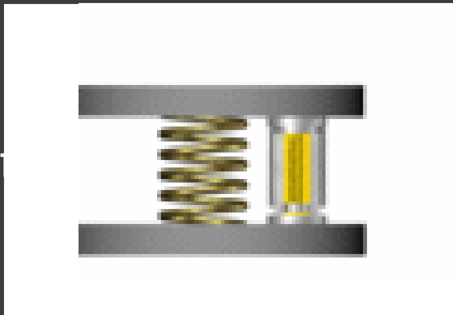
A1: THz frequencies and picosecond time scales are ubiquitous in nature.

- Small molecule rotations
- Biologically-important collective mode vibrations of proteins.
- Electron spin resonances
- Absorption in polar liquids due to frustrated rotation and collective modes.
- Collisions times of gas molecules at room temperature.
- Resonant frequencies of electrons in semiconductors and their nanostructures
- Superconductor gap energies.
- Scattering rates of electrons in correlated metals.
- Phonon frequencies in molecular crystals.
- Electron transit times in *Intel's* new THz transistor.

A2: Low temperature → an interesting limit is ω>T

10 K ~ 200 GHz → THz is higher than T (quantum limit
high (quantum [illegible]s)

Important information gained by driving a system at its natural frequencies



MANY MORE!

Problems with accessing THz.

Although incredibly useful, working in this regime has been difficult.

<u>Traditional paucity of strong (enough) sources, sensitive detectors, measurement configurations compared to other frequencies.</u>

- **Bottom up** (electronics: wires etc.)
- **Top down** (photonics: lenses, mirrors etc.).

- Pushing up from low frequencies → measure configuration starts to 'broadcast'.

- Pushing down from high frequencies → Signal is swamped by background (Wiens law 10K → 1THz). Hard to separate signal from noise without cryogenic detectors and environment. Diffraction.

THz motivation for strongly correlated system

Correlated system → large ensemble of strongly interacting, but fundamentally simple particles acting collectively to exhibit complex emergent quantum phenomena. Ex. superconductors, magnets, etc.

Not necessarily reducible to the short distance atomic scale… 10^{24} particles acting collectively to give qualitatively different physics at long length and time scales…

We want to probe scales which are long, but not ***so*** long… and short, but not ***so*** short… intermediate scales

Important frequency scale tends to be one of order where the temperature are exhibited.   Ex. superconductor $2\Delta/kT_c$.

1 THz ~ 48 Kelvin ~ 4 meV

Conclusions…

-Discussed our evidence for a topological phase transition in topological insulators

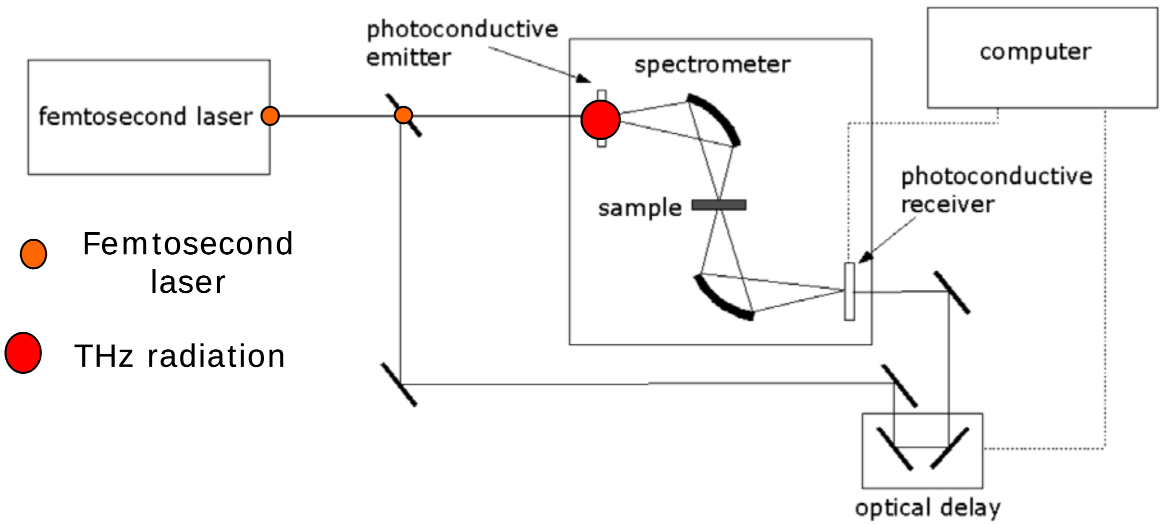
-Demonstrated novel “kink” excitations in linear spin-chains

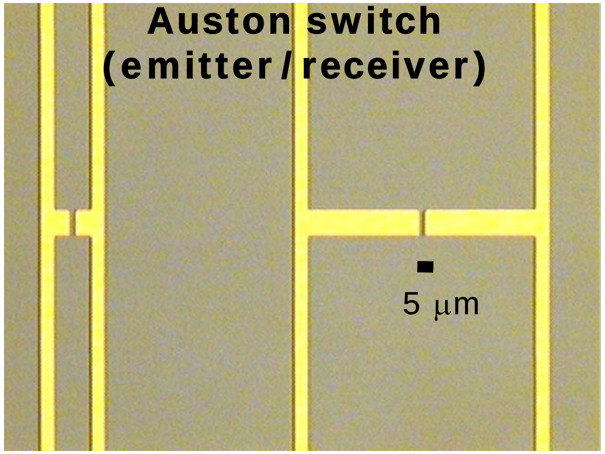
-Evidence string excitations in spin ice?

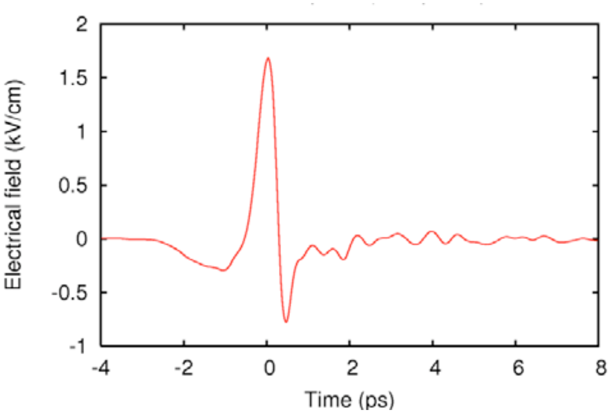
-Presented evidence for anisotropic conductivity in cuprate superconductors (nematicity? mangetoelectricity?)

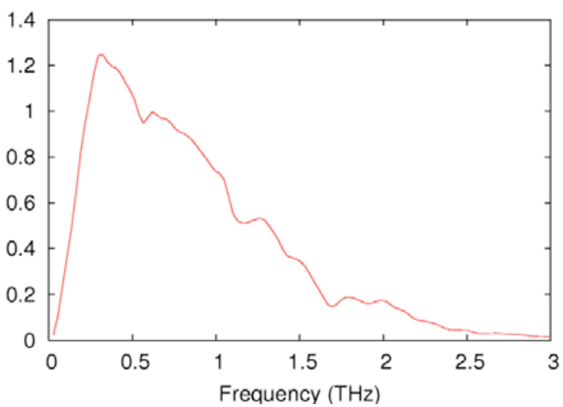
…all the while emphasizing the role that new technology plays in our ability to make measurements

THz Time Domain Measurements





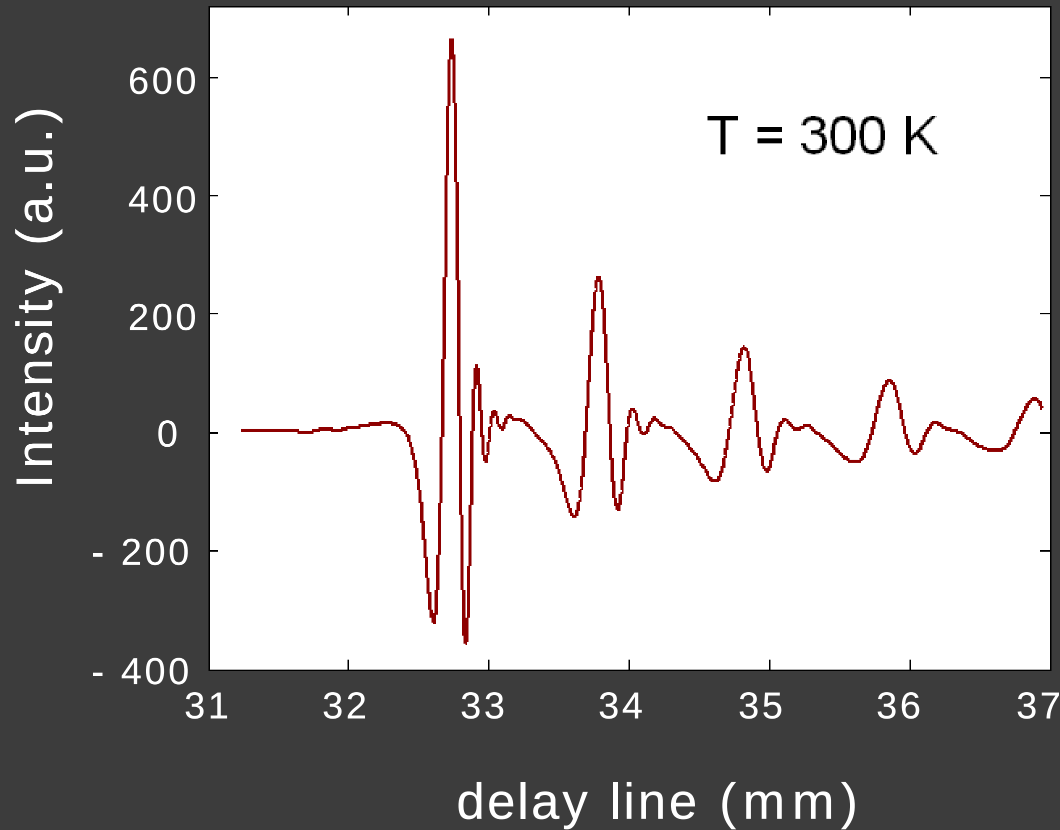




$$T(\omega) = \frac{4n}{n+1}\frac{e^{i\Phi_s}}{n+1+\sigma(\omega)dZ_0}$$

- Femtosecond laser pulse excites photoconductive emitter and receiver
- Coherent detection of field allows **complex** response to be measured. 100 GHz - 3 THz, 1.5K - 300K.
- Can measure complex response of charge and spin with high single to noise and very high energy resolution <5μeV.
- Can be complementary to inelastic neutron scattering

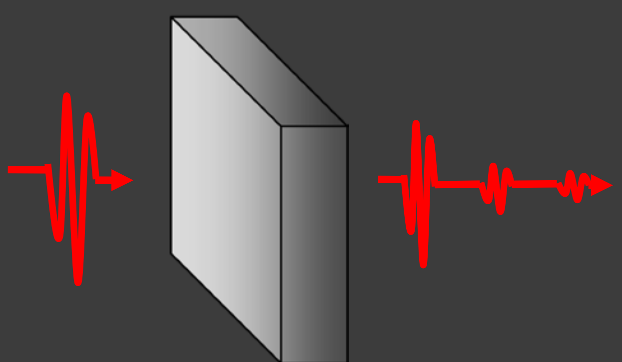
THz time domain measurements



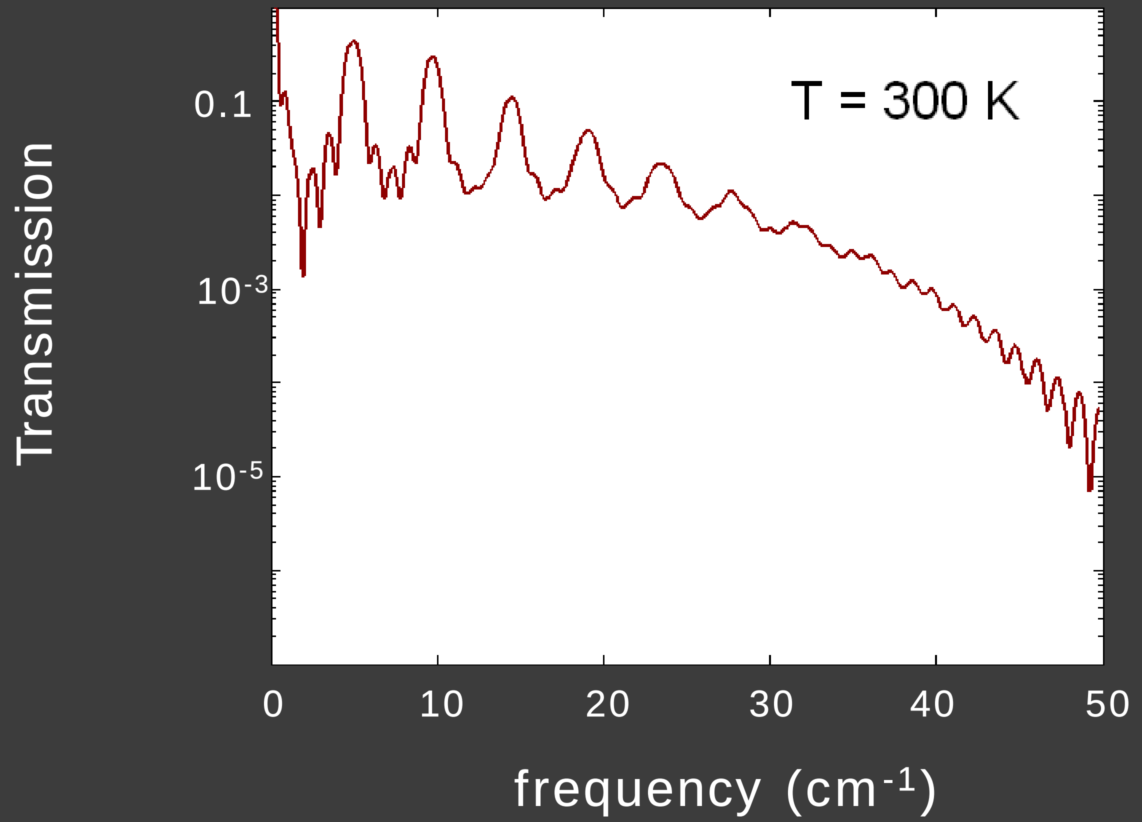
cooling down

warming up

- Time domain transmission of 60 μm thick $SrTiO_3$
- $SrTiO_3$ is a quantum paraelectric. ~40K ferroelectricity destabilized by zero point motion. ε ~ 40,000 at 4K.



THz time domain measurements

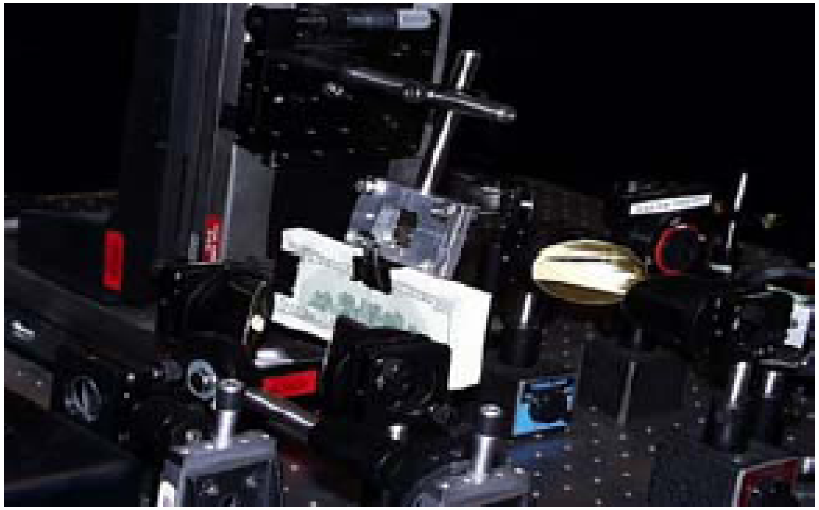


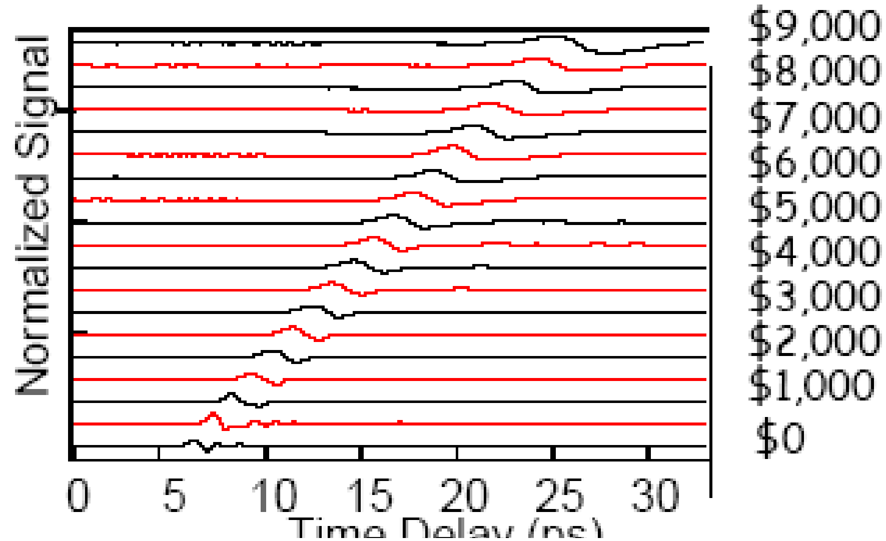
cooling down

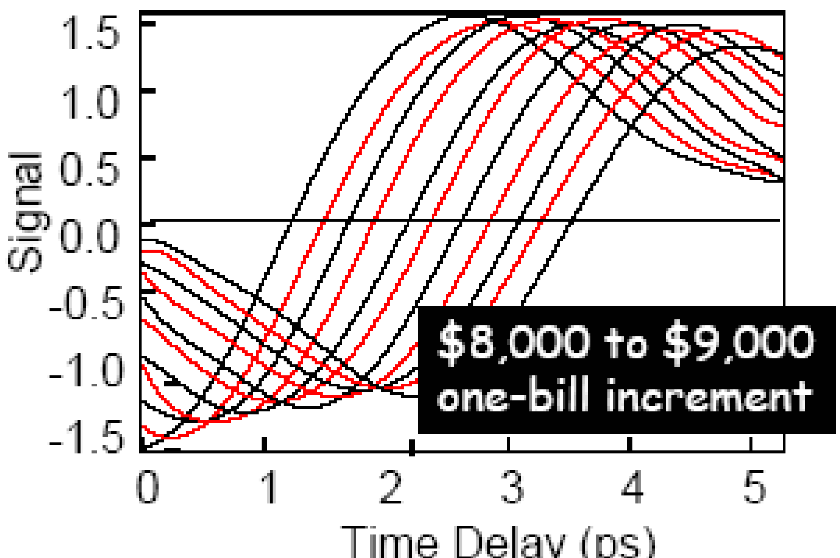
warming up

- Frequency domain transmission spectrum of 60 μm thick pure $SrTiO_3$
- *ps* pulse has THz Fourier components in it.

Counting $$$ @ the speed of light







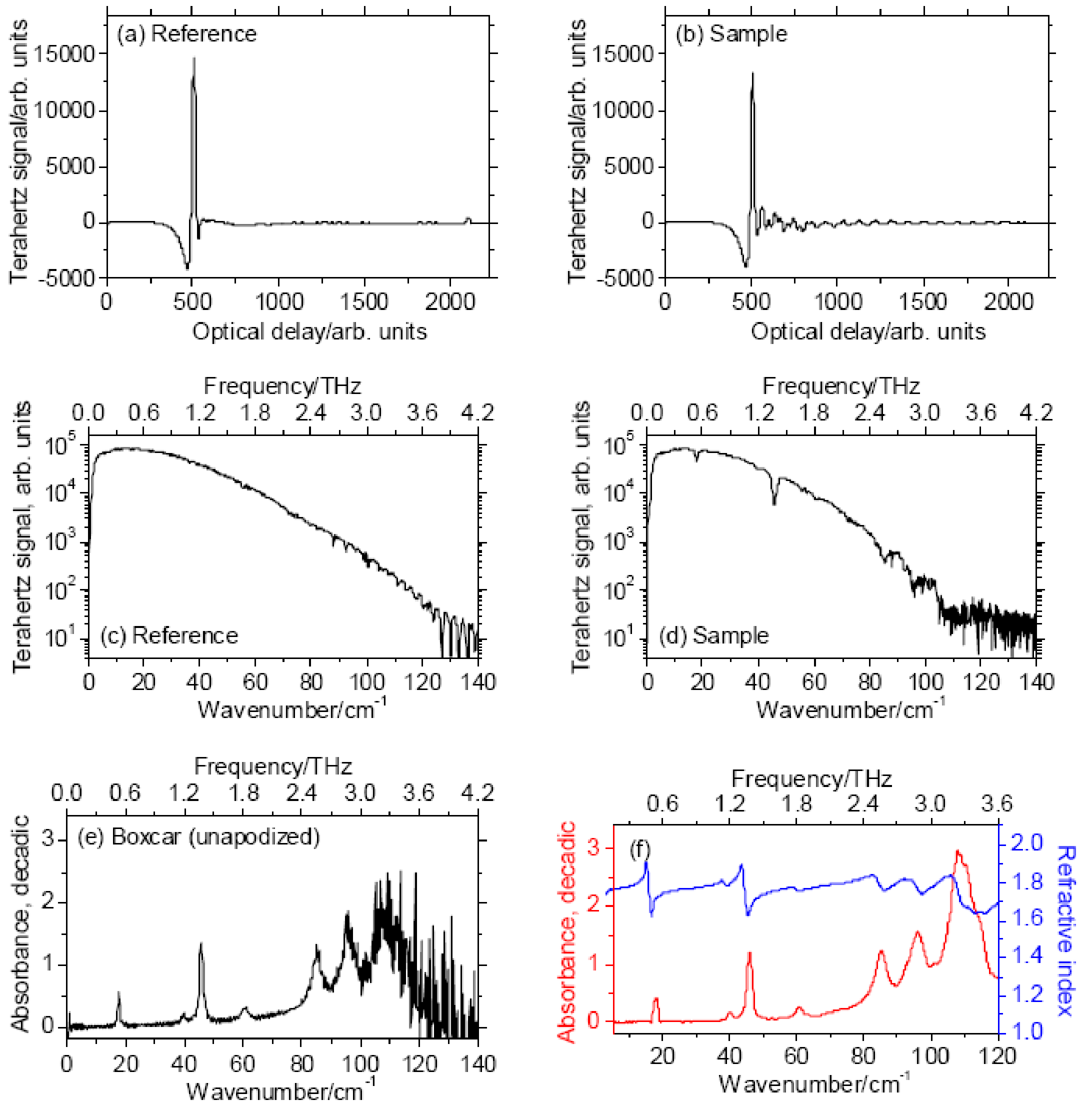
Time IS Money!

THz Time Domain Measurements

Transmission of time limited pulse

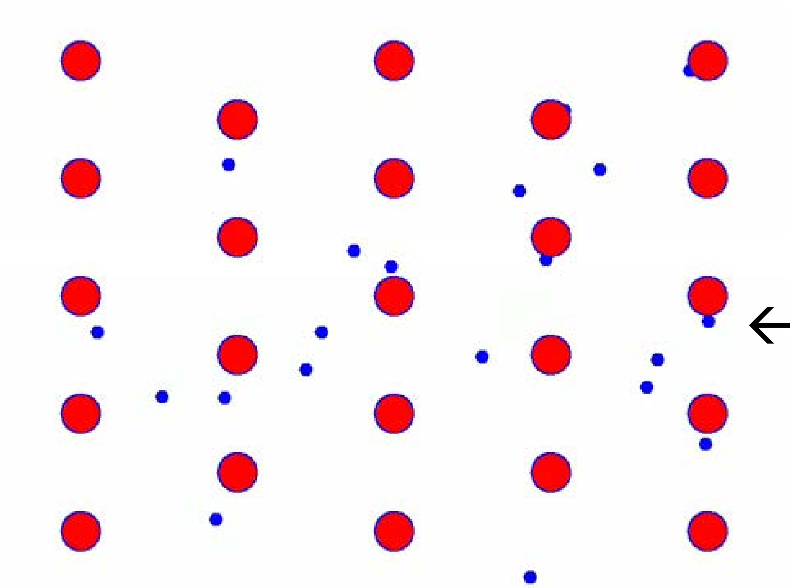
Fourier Transform

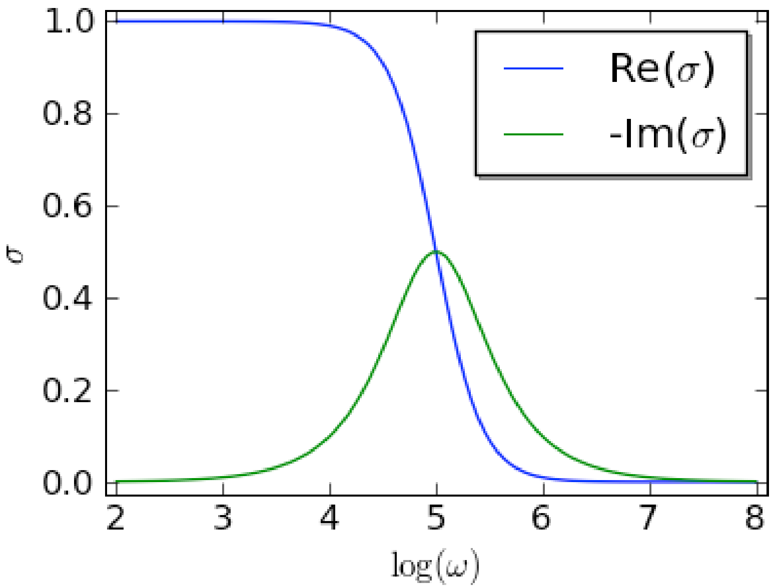
Given both transmitted amplitude and phase, any set of optical constants can be found.



Lactose Absorption Spectra
NPA *unpublished*

Drude conductivity





$$mx'' = -eE - mx'/\tau$$

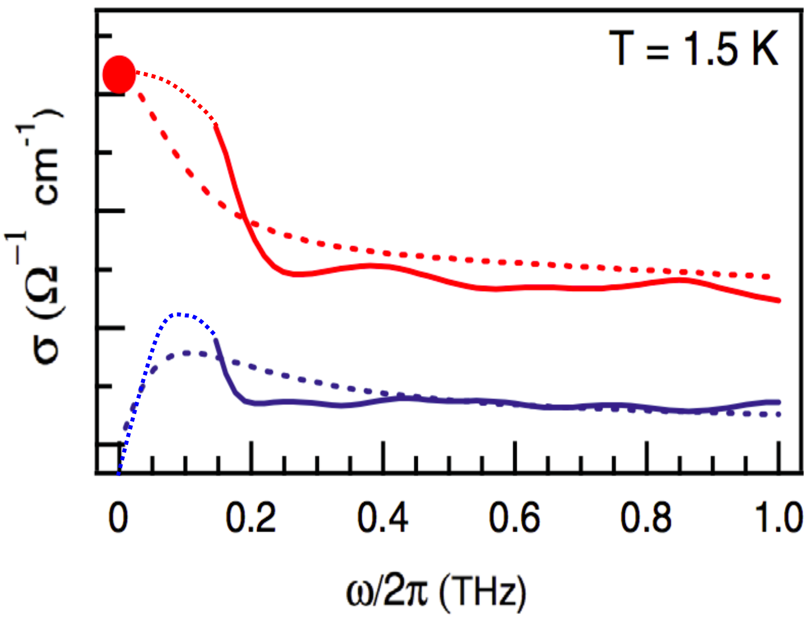
$$J = \sigma E$$

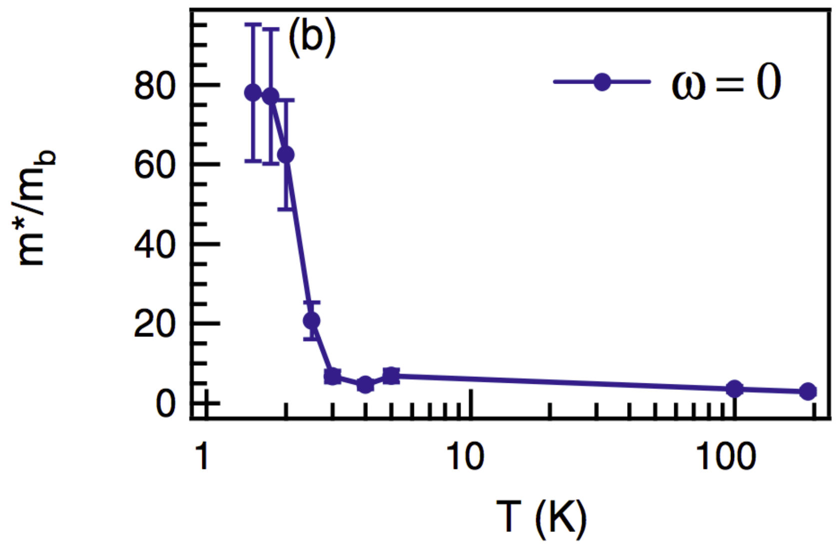
$$\sigma(\omega) = \frac{Ne^2\tau}{m}\frac{1}{1-i\omega\tau} = \frac{Ne^2\tau}{m}\frac{1+i\omega\tau}{1+\omega^2\tau^2}$$

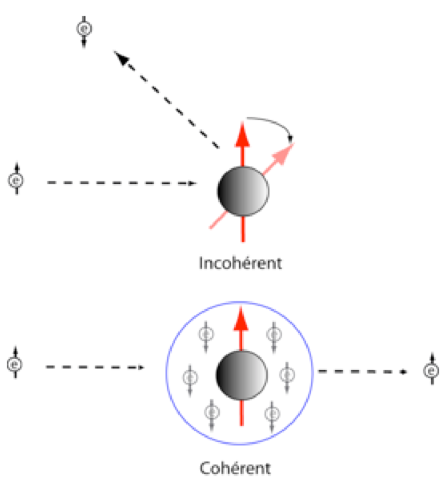
PHYSICAL REVIEW B **85**, 155105 (2012)

Low energy electrodynamics of the Kondo-lattice antiferromagnet $CeCu_2Ge_2$

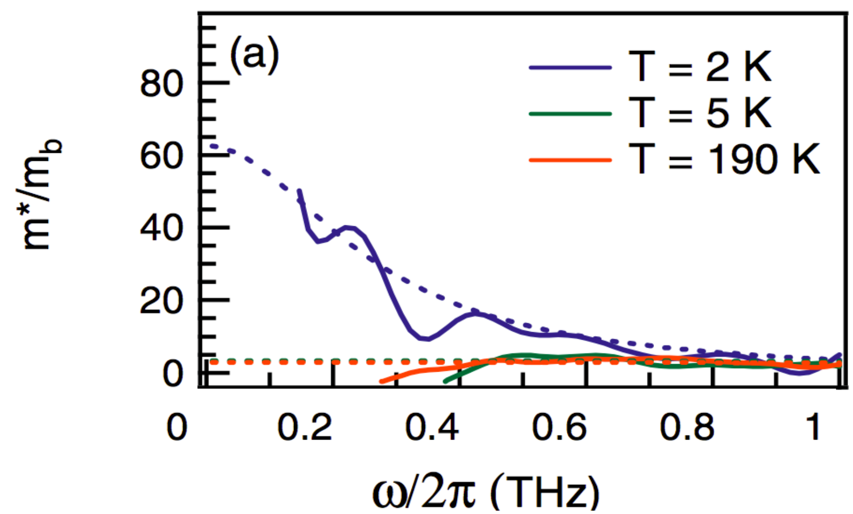
G. Bossé,[1] L. S. Bilbro,[1] R. Valdés Aguilar,[1] LiDong Pan,[1] Wei Liu,[1] A. V. Stier,[1] Y. Li,[2] L. H. Greene,[2] J. Eckstein,[2] and N. P. Armitage[1]







Electron-spin interaction gives greatly enhanced mass through coherent Kondo effect

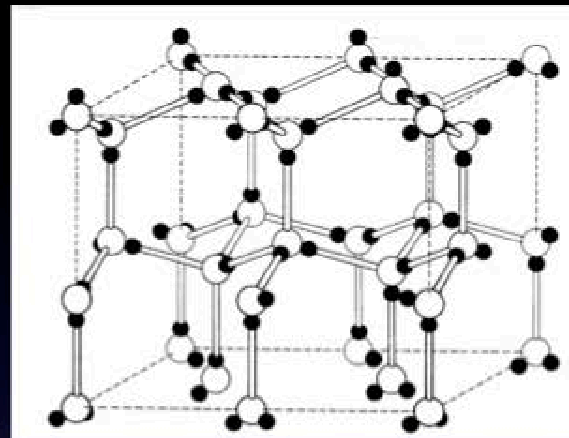


Transport in topological insulators

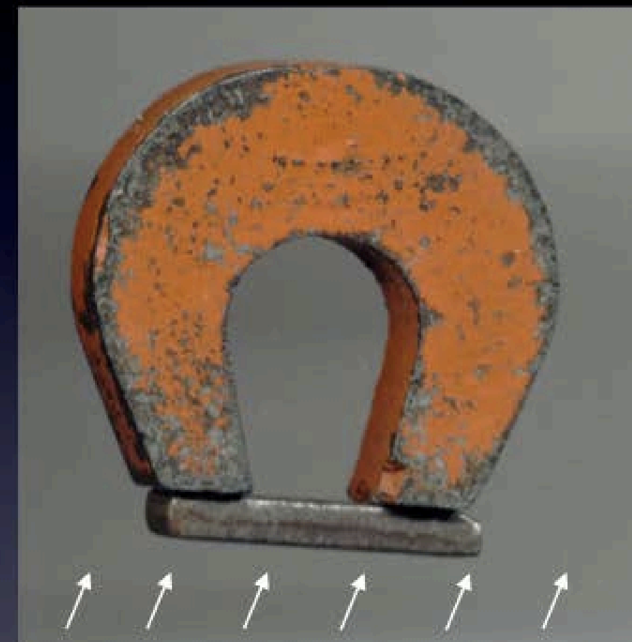
Proposed "Flipper Bridge" between Hong Kong (left-hand traffic) and mainland China (right-hand traffic). Design by nl architects (www.nlarchitects.nl). Concept by Phillip Hoffman, Aarhus University

Ordered States of Matter

Low temperature state has **less** symmetry than high temperature state (translational, rotational, gauge symmetry broken)

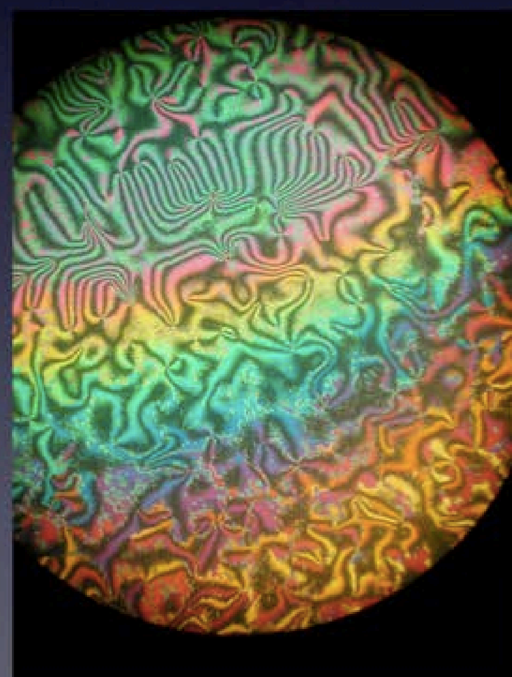


crystals

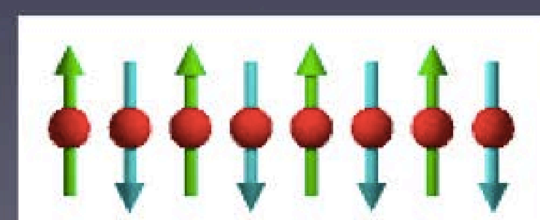


ferromagnetism

superconductors

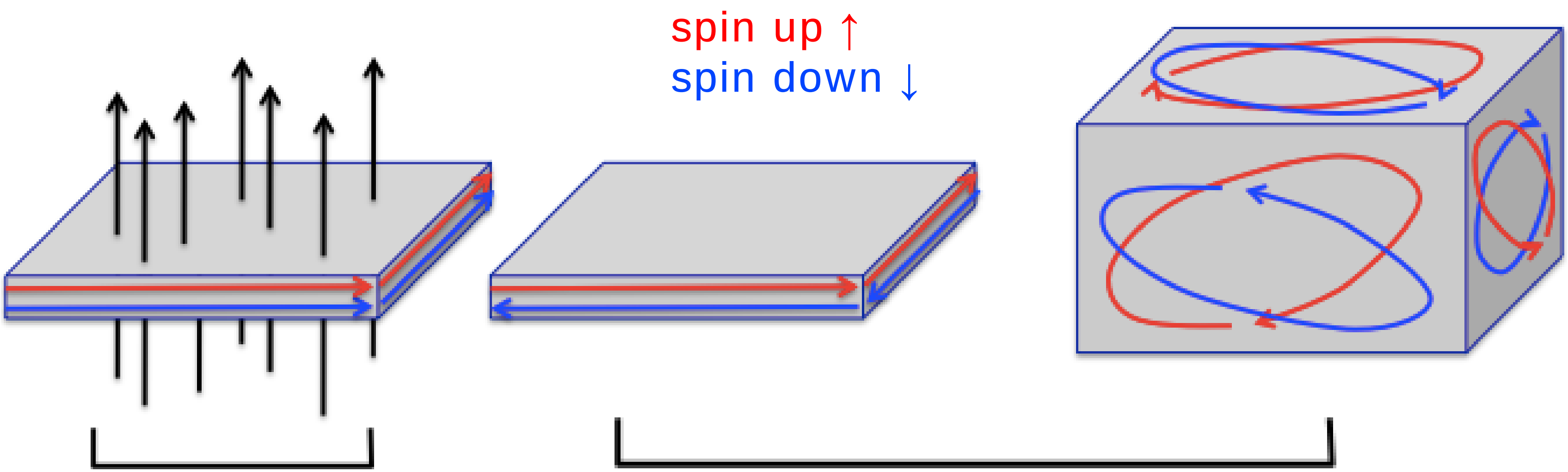


liquid crystals



antiferromagnetism

Topological States of Matter → Distinguished not by broken symmetries, but "topological structure" of wavefunctions



Quantum Hall

2D TI

3D TI

2D metal in large B field

Exact quantization of Hall resistance in units of h/ne^2

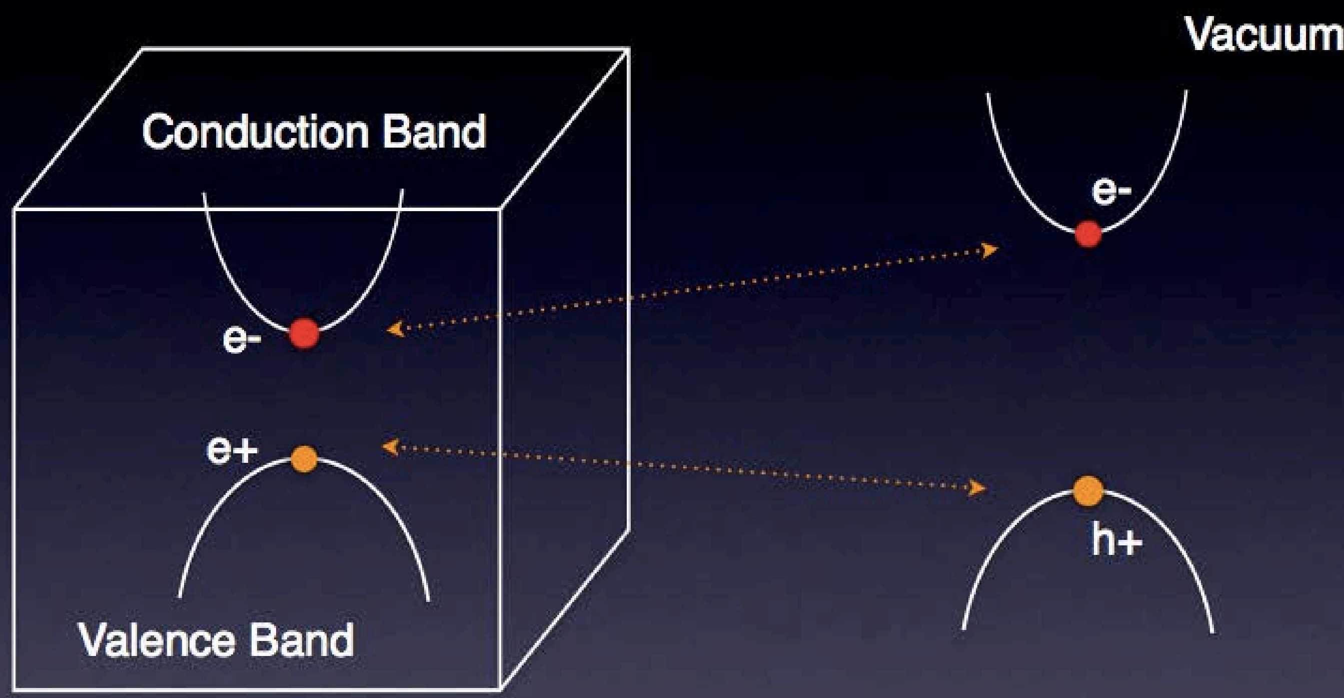
Broken time-reversal symmetry

time-reversal symmetric

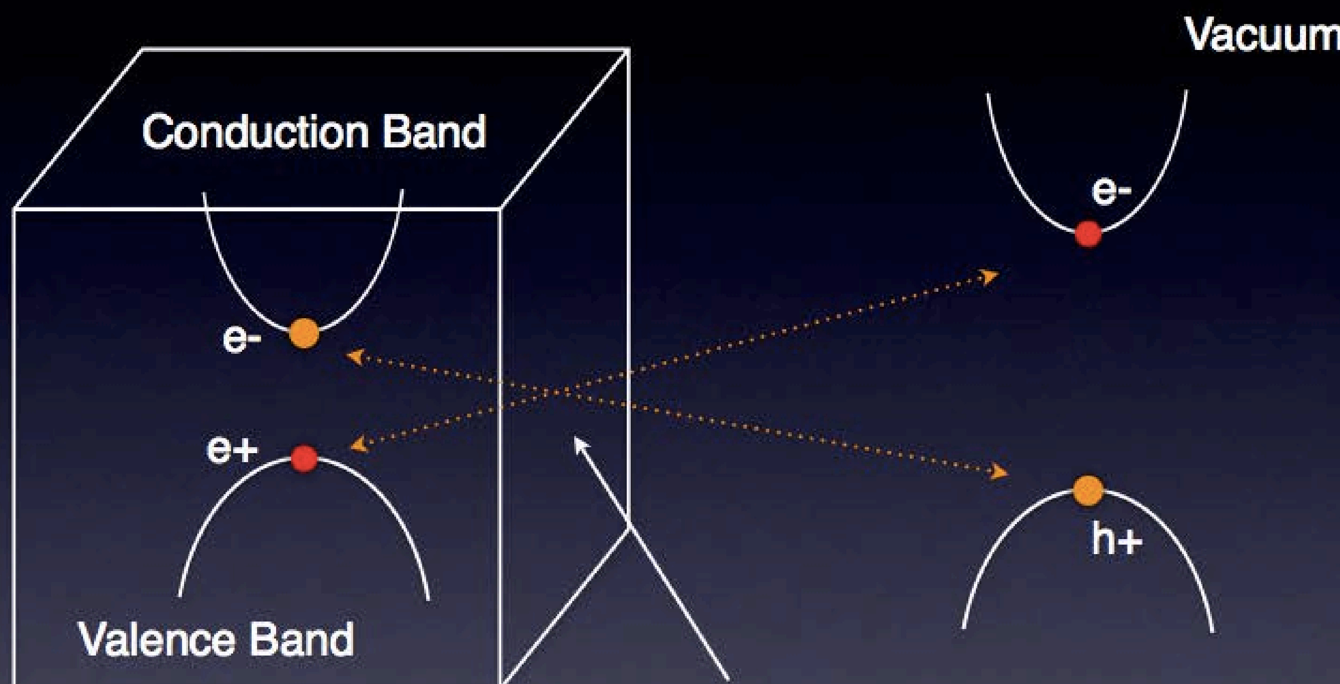
Role of B field is played by spin-orbit coupling→

$$H_{SOC} = \frac{\hbar}{4m^2c^2}(\nabla V \times \vec{p})\vec{\sigma}$$

Conventional Insulator: Adiabatically connected to the vacuum



Topological Insulator: Adiabatically disconnected from the vacuum



Gap closes between two different vacua; Topologically protected surface metals

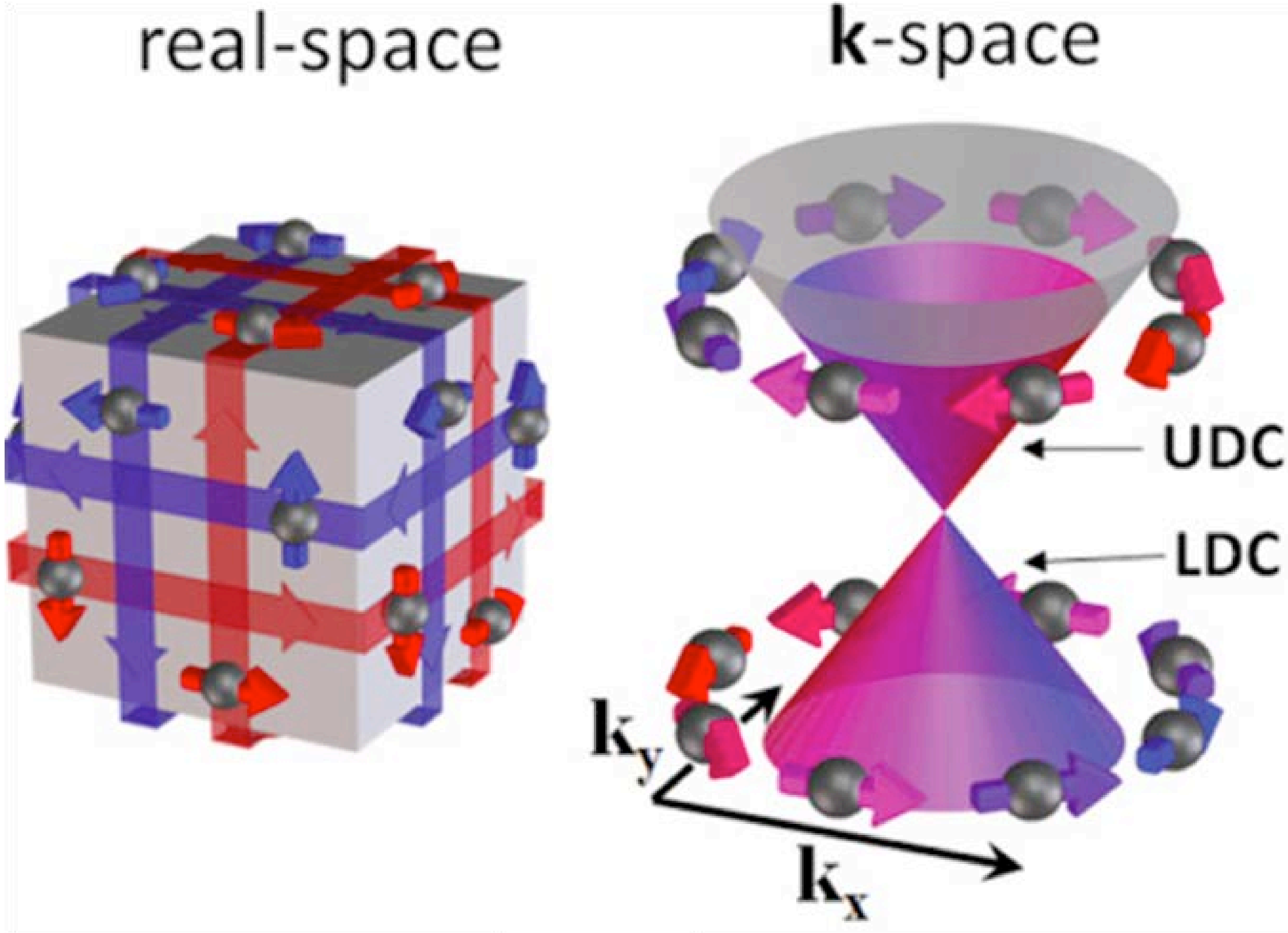
Theory --> Fu, Kane, Mele, Zhang, Qi, Bernevig, Hughes, Roy, Moore, Balents, Franz, others?; Experiment --> Molenkamp, Hasan, many others

Driven by spin-orbit coupling --> heavy atom compounds

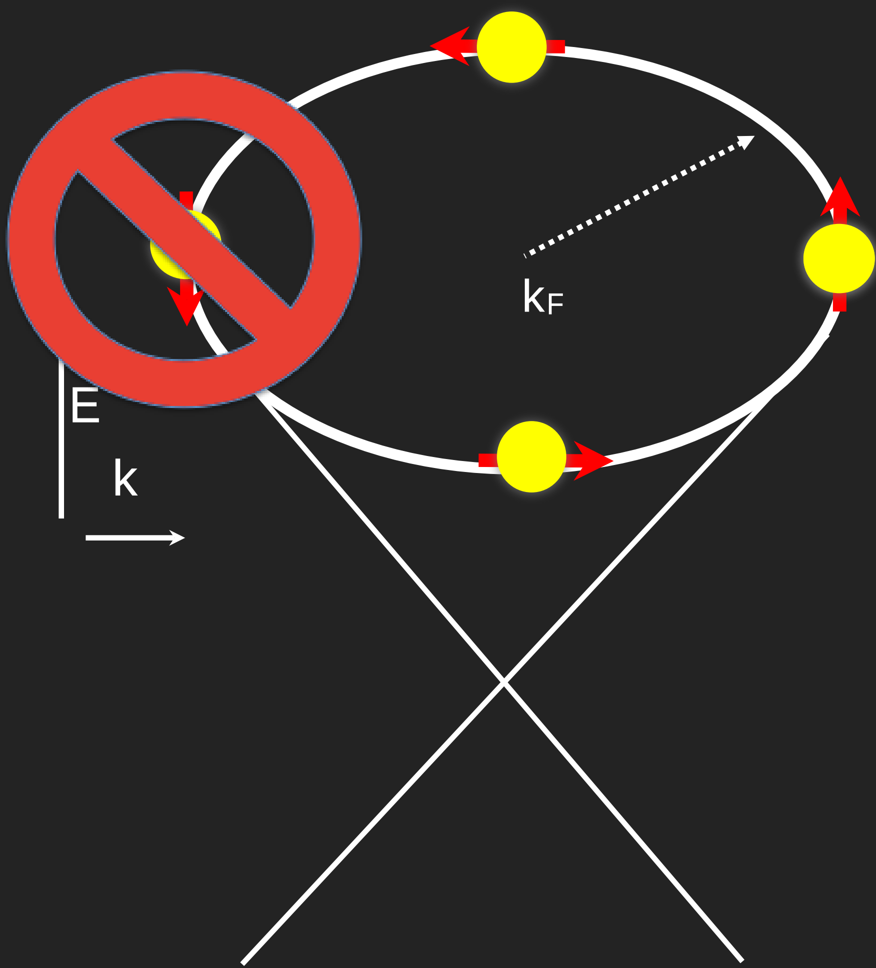
$$H_{SOC} = \frac{\hbar}{4m^2c^2}(\nabla V \times \vec{p})\vec{\sigma}$$

Transport in topological insulators

Proposed "Flipper Bridge" between Hong Kong (left-hand traffic) and mainland China (right-hand traffic). Design by nl architects (www.nlarchitects.nl). Concept by Phillip Hoffman, Aarhus University


real-space
k-space
UDC
LDC
k_y
k_x

Topological protection in transport



Small angle scattering does not usually affect transport lifetime. Backscattering dominates

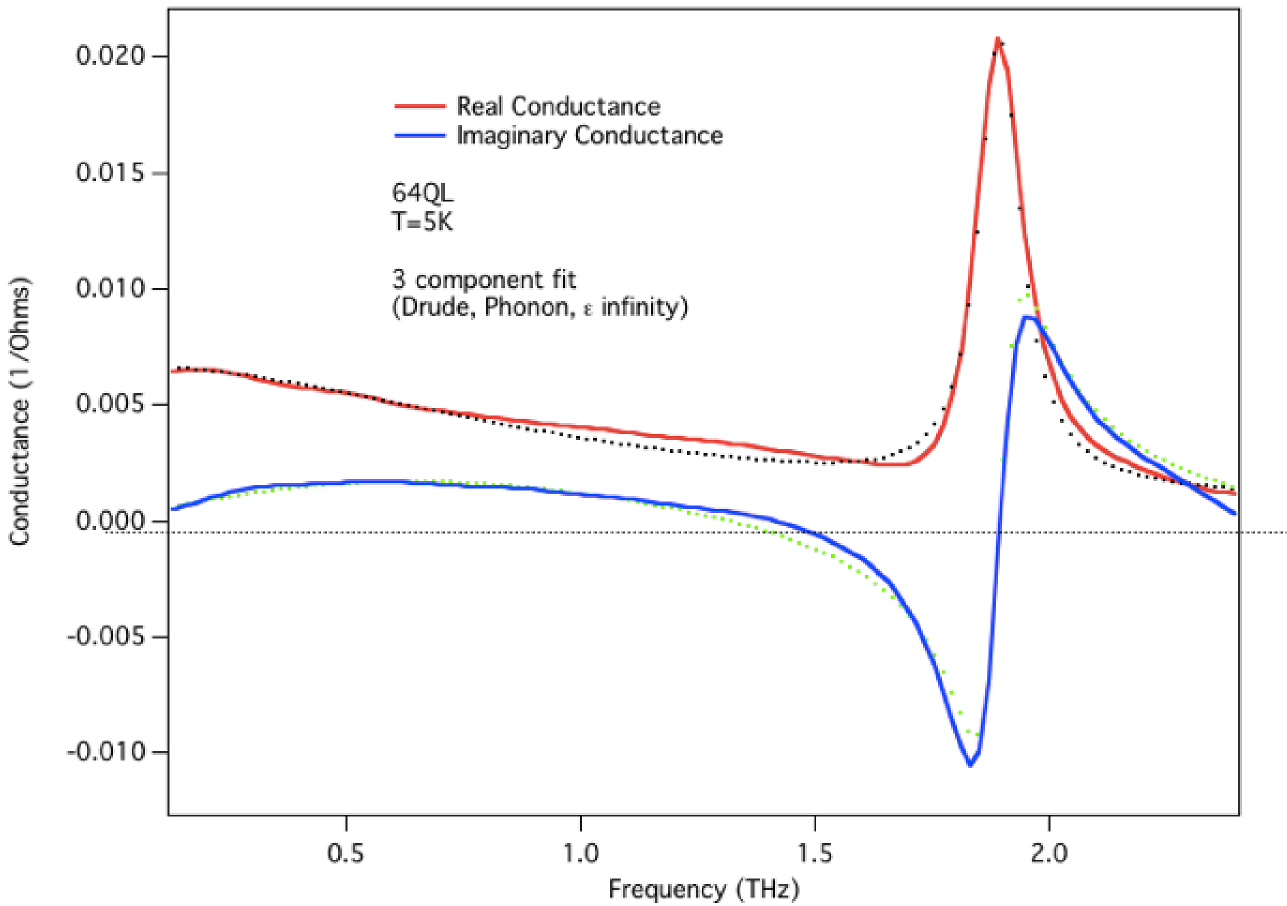
$$1/\tau_{trans} = (1/\tau_{qp})\frac{1}{2\pi}\int d\theta\left(\frac{1-\cos\theta}{2}\right)$$

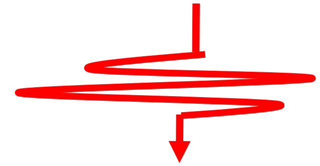
$$1/\tau^{TSS}_{trans} = (1/\tau_{qp})\frac{1}{2\pi}\int d\theta\left(\frac{1-\cos\theta}{2}\right)\left(\frac{1+\cos\theta}{2}\right)$$

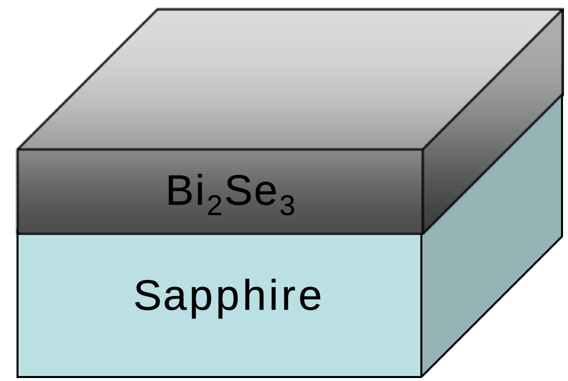
In TI SS 180º backscattering suppressed → "spin"-momentum locking

Differences of order unity in classical transport regime
→ small but significant

THz Conductance Measurements in Bi_2Se_3 films







MBE grown films

Surface conduction dominates for less than 150 unit cells (QL)

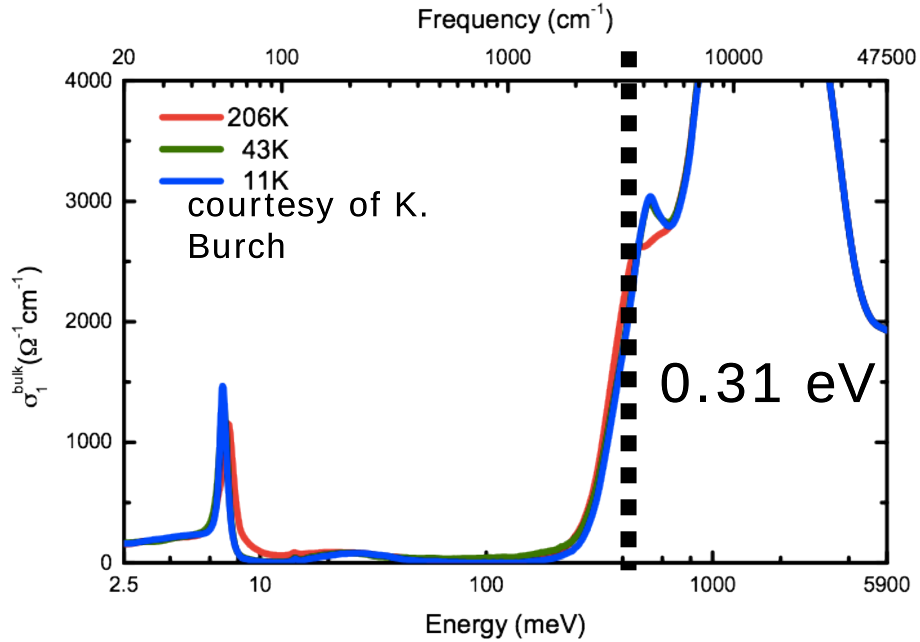
Conductance is a complex quantity

$E\sigma = J$

$$T(\omega) = \frac{4n}{n+1}\frac{e^{i\Phi_s}}{n+1+\sigma(\omega)dZ_0}$$

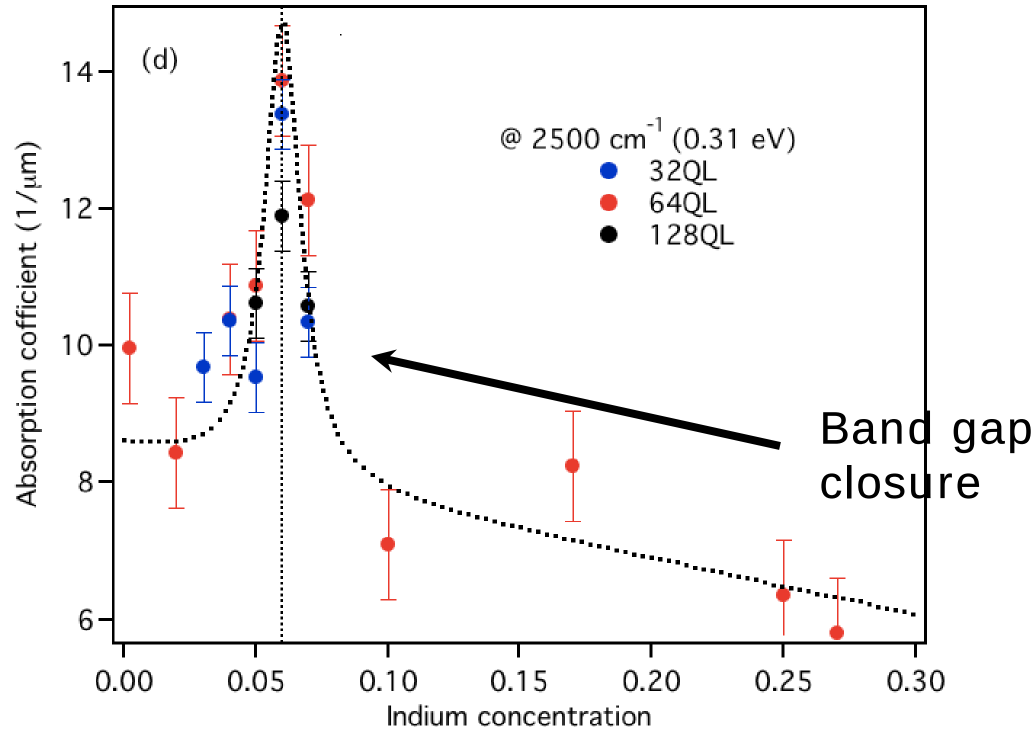
Topological Transition in $Bi_{2-2x}In_{2x}Se_3$

Dissipative conductivity over large energy range for pure x=0 Bi_2Se_3

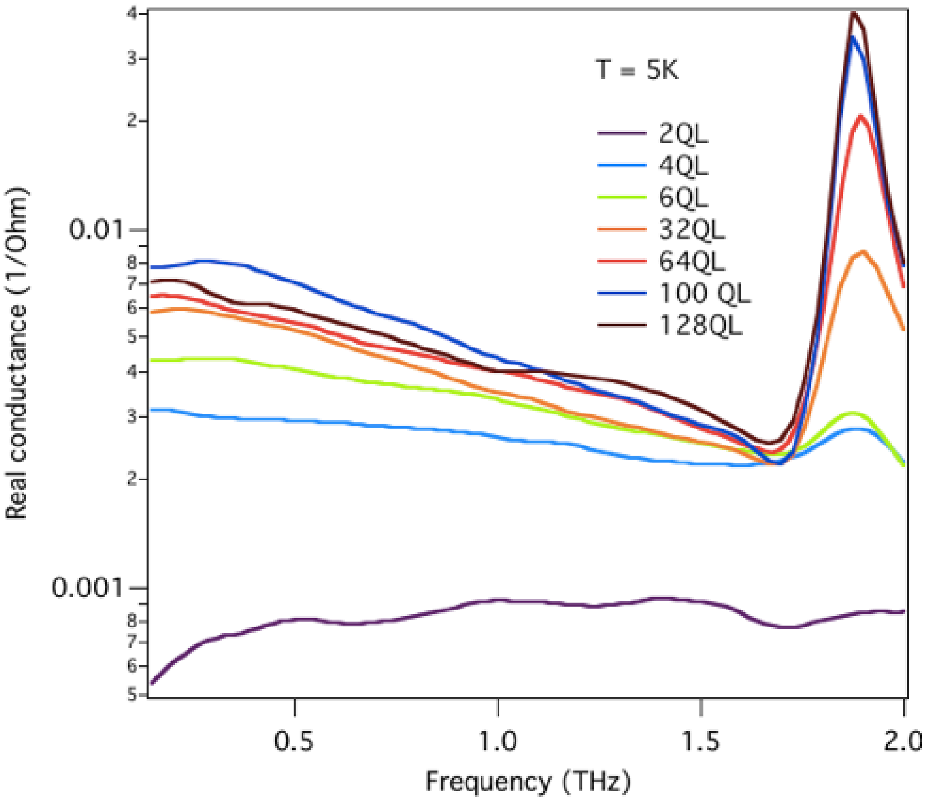


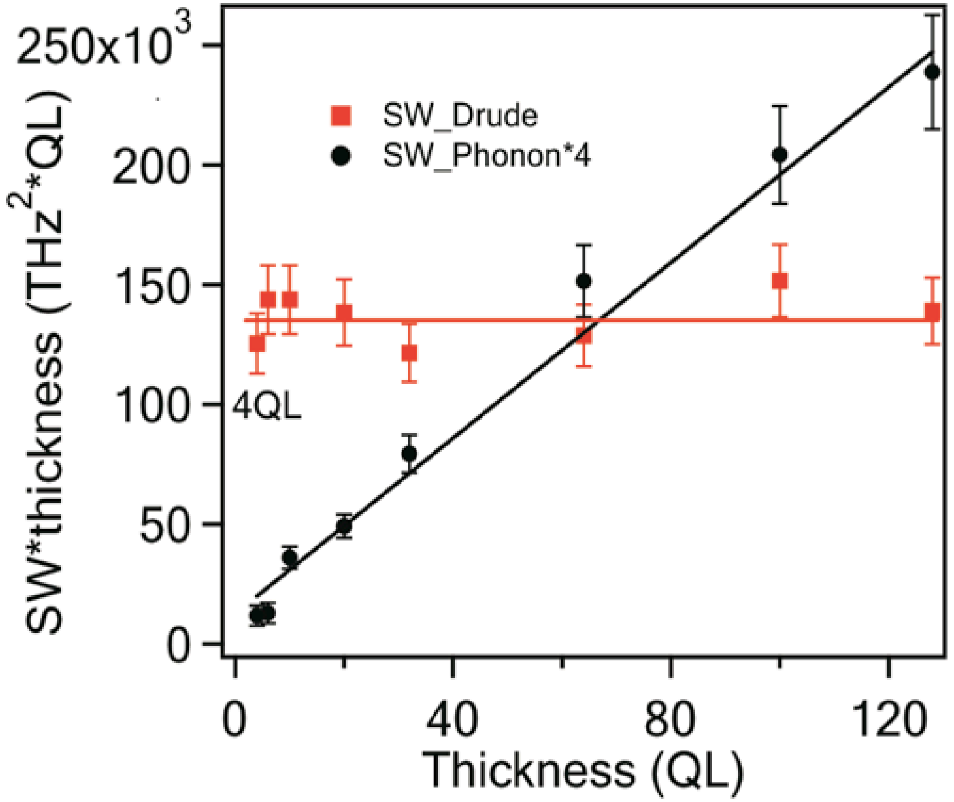
In_2Se_3 is a band insulator with same structure

With In doping bulk band gap closing correlated with low energy metallic response



Thickness independent transport in Bi_2Se_3 films





Important quantities are areas and widths of peaks -- We fit them

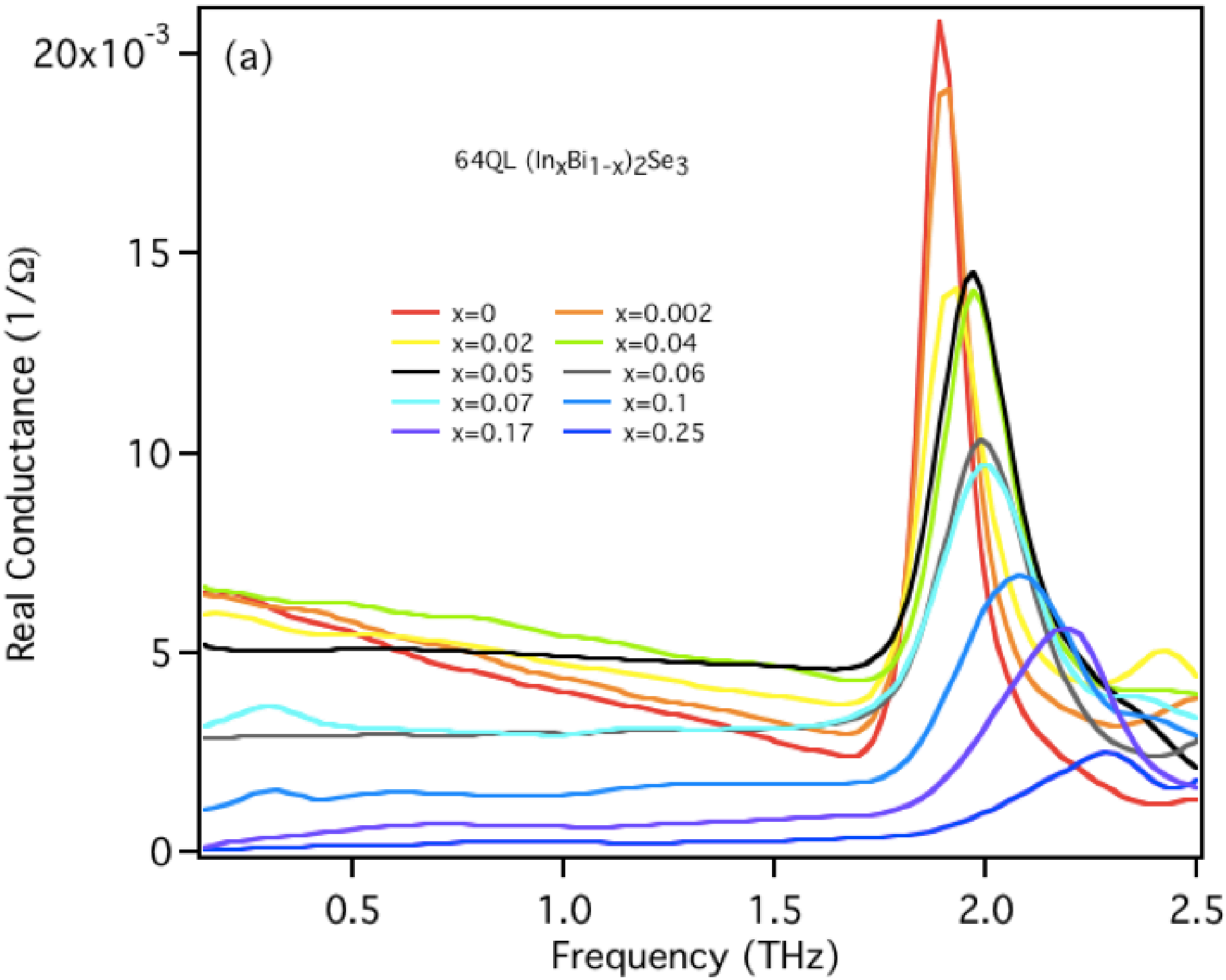
$$8\int_0^W \sigma_1(\omega)d\omega = \frac{4\pi ne^2}{m_b}$$

Conductivity Sum Rule

Scattering rate of Drude piece dependent on thickness, but “area” is preserved.

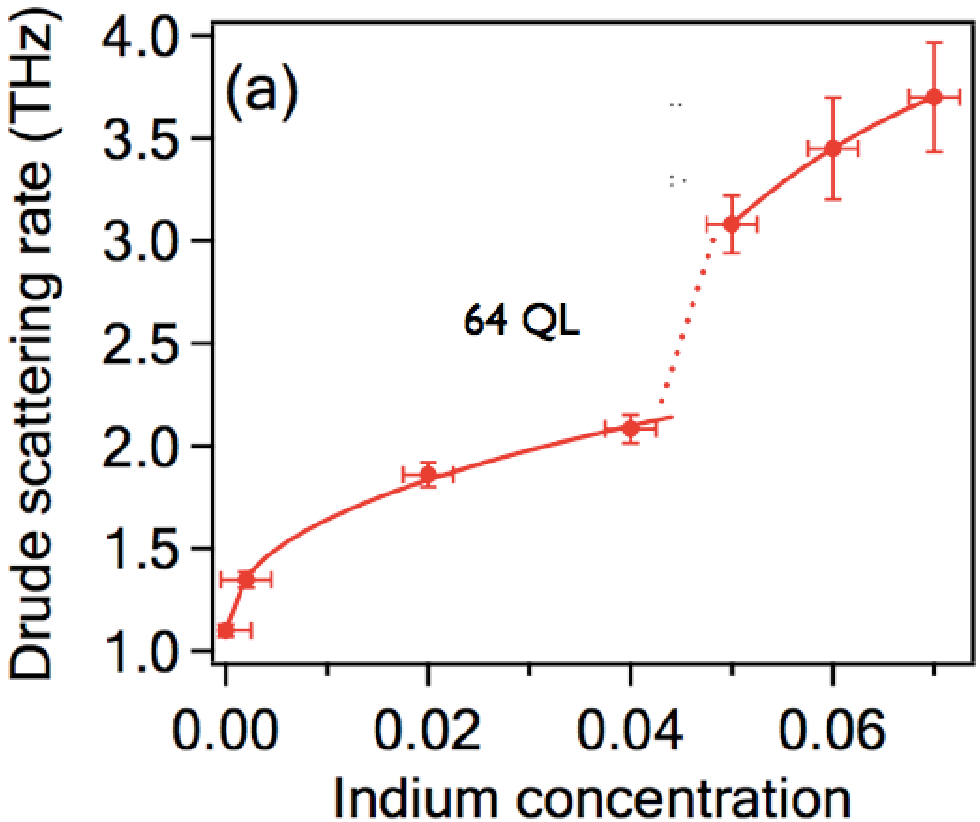
Area of phonon component depends linearly on thickness

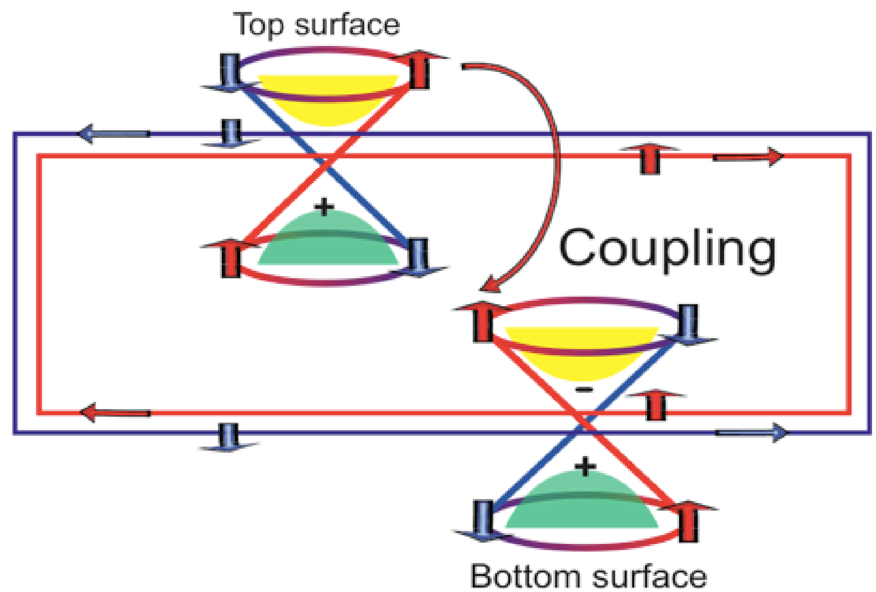
Topological Transition in $Bi_{2-2x}In_{2x}Se_3$



Band gap inversion (transition to normal insulator) happens at 6% doping

Topological Transition in $Bi_{2-2x}In_{2x}Se_3$





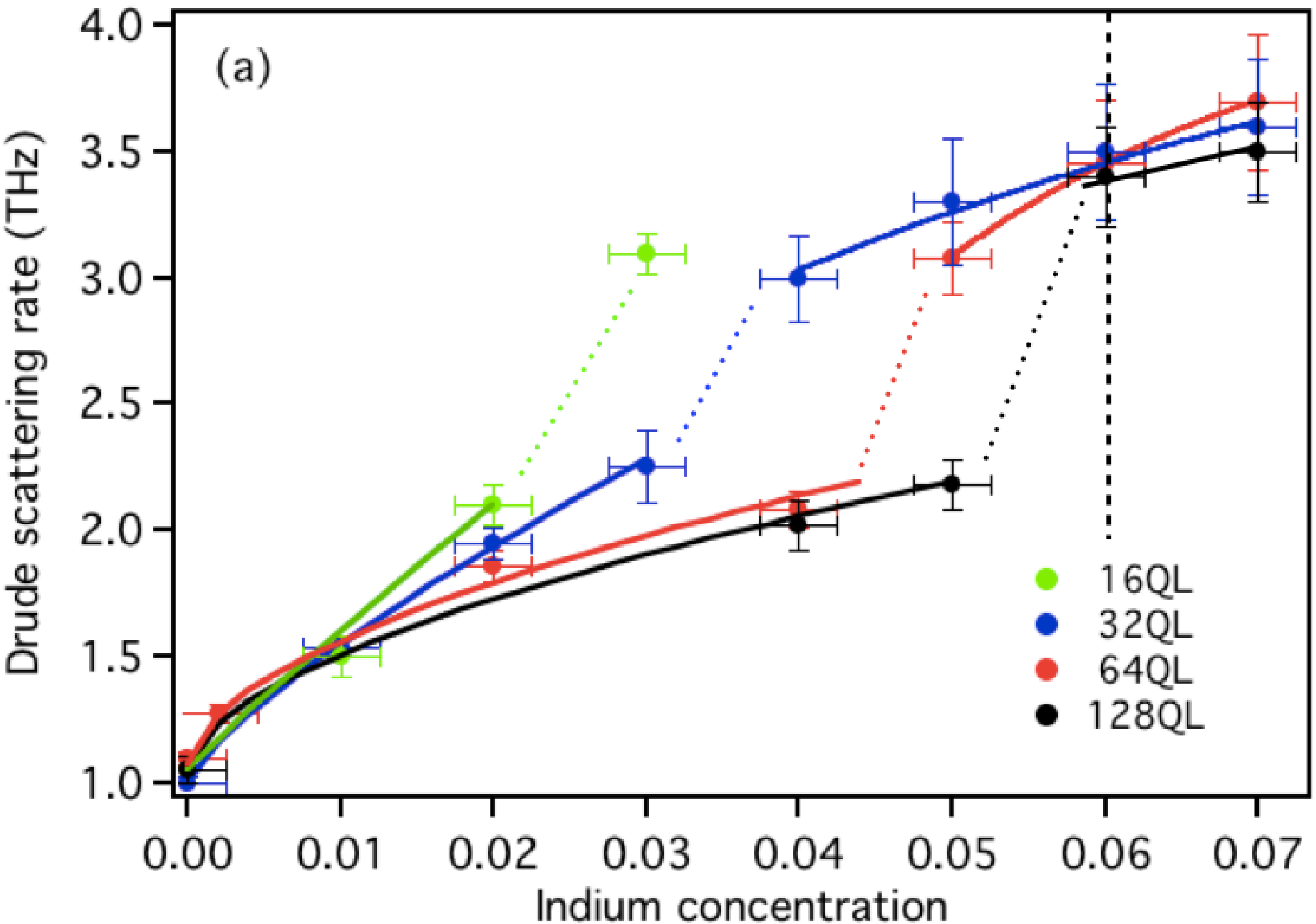
$\xi \sim v_F/\Delta \sim 2$ nm * 0.3 eV/$\Delta(x)$

Penetration depth of surfaces is governed by band gap

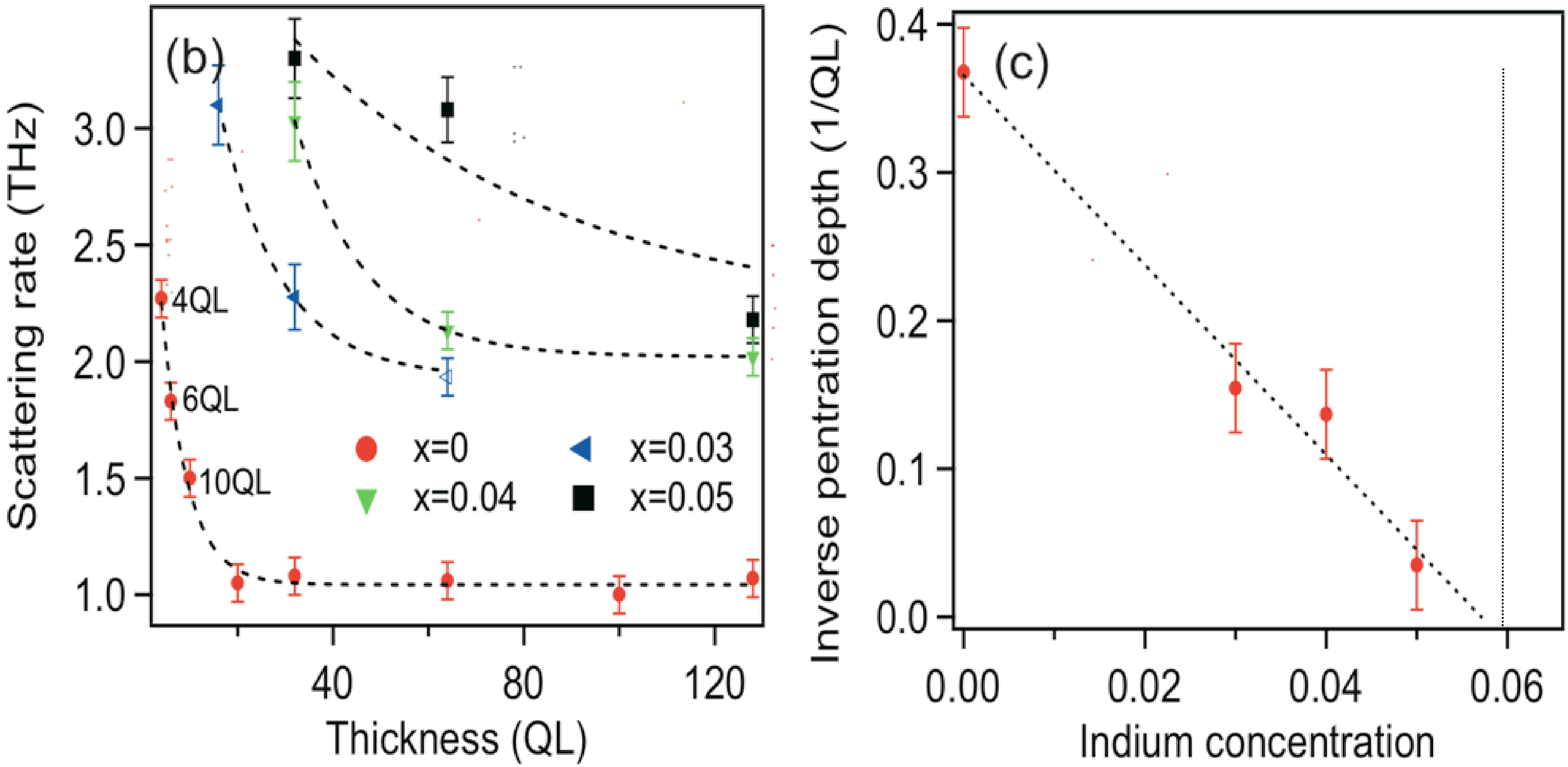
Enhanced scattering "near" the TI/nTI transition

But wait! ... there is a mismatch between x~0.05 where jump in $1/\tau$ is seen and x~0.06 where band gap closes

Scattering rates through the TI/non-TI “phase transition”



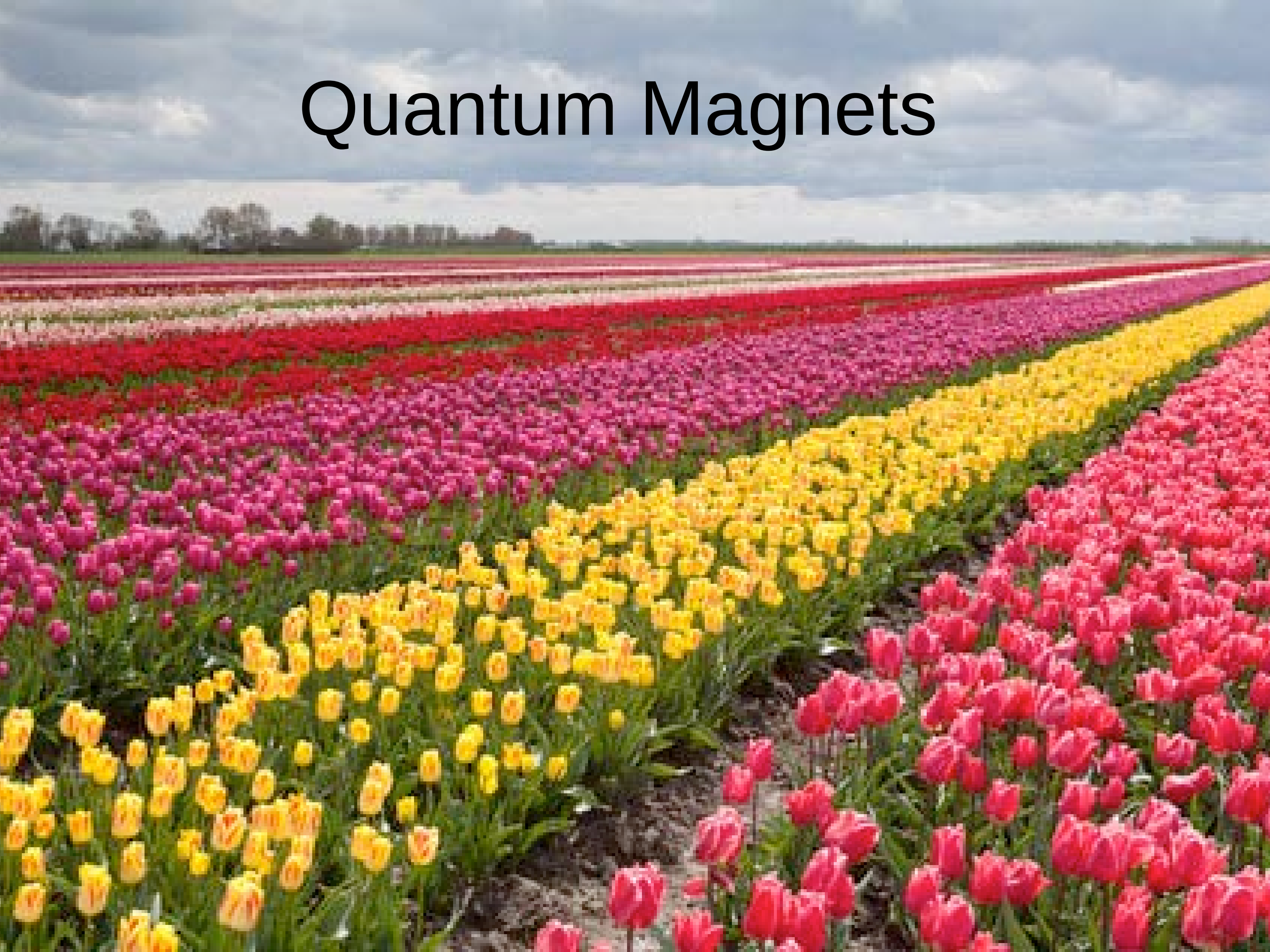
Finite size scaling analysis



Finite size analysis gives dependence of critical length

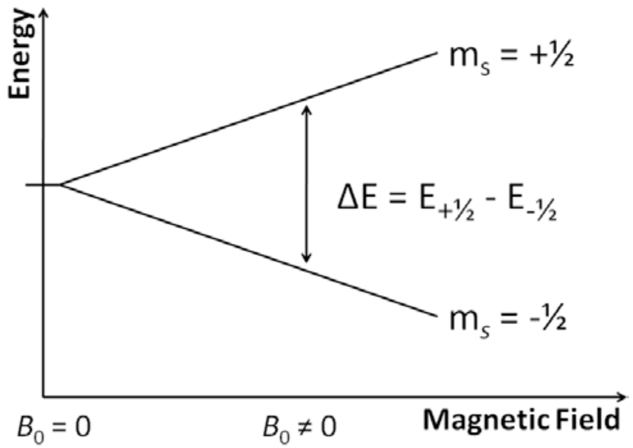
Evanescent length of surface state plays role or correlation length of phase transition

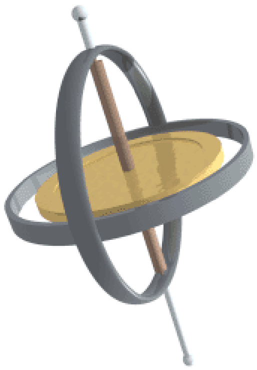
Quantum Magnets



THz broadband electron spin resonance (ESR)

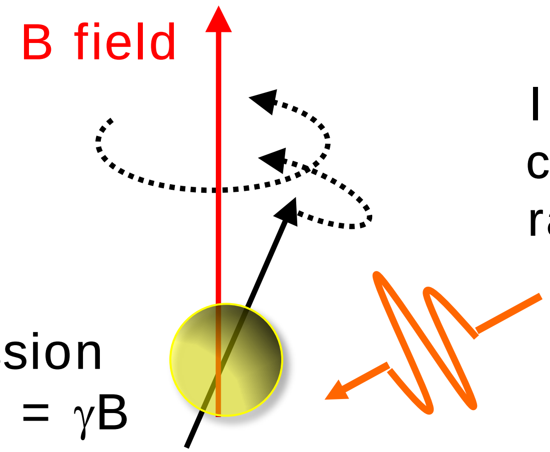
Excite **magnetic** dipole with light's time varying magnetic field





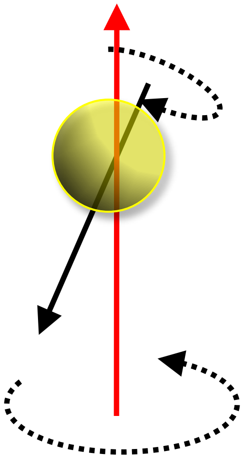
In transmission

$\ln(T(\omega)) = -\omega\chi(q \to 0, \omega)$

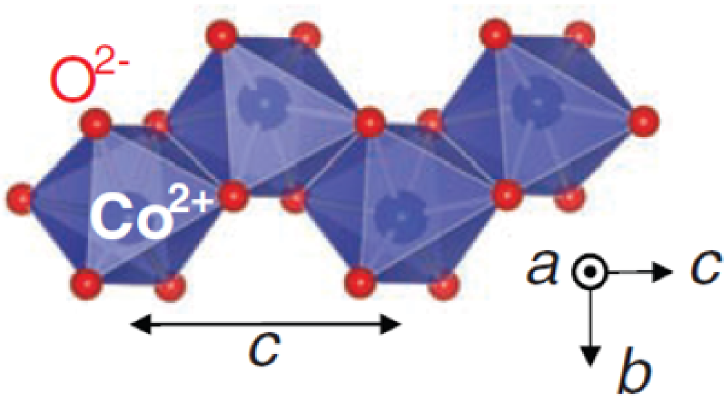


Precession with $\omega = \gamma B$

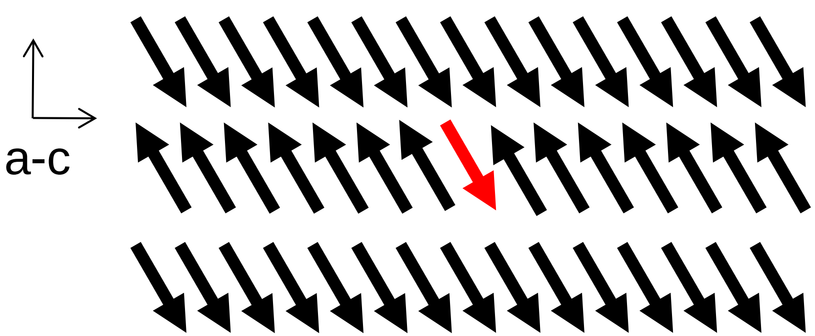
Illumination with ω circularly polarized radiation



$CoNb_2O_6$ – Ising chain material



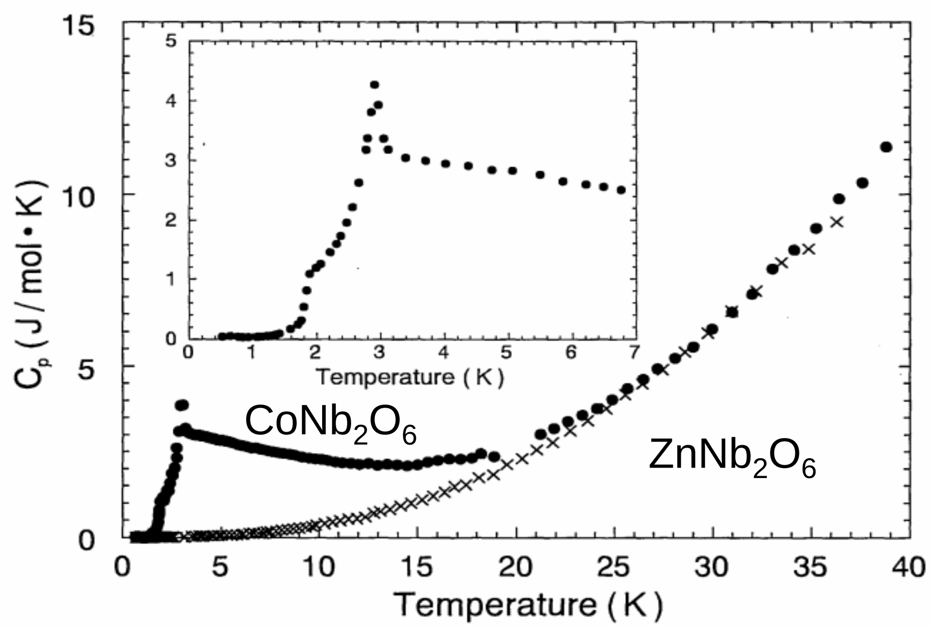
R. Coldea et. al., Science **327**, 177 (2010)
E8 structure in transverse field



Strong Ising interactions

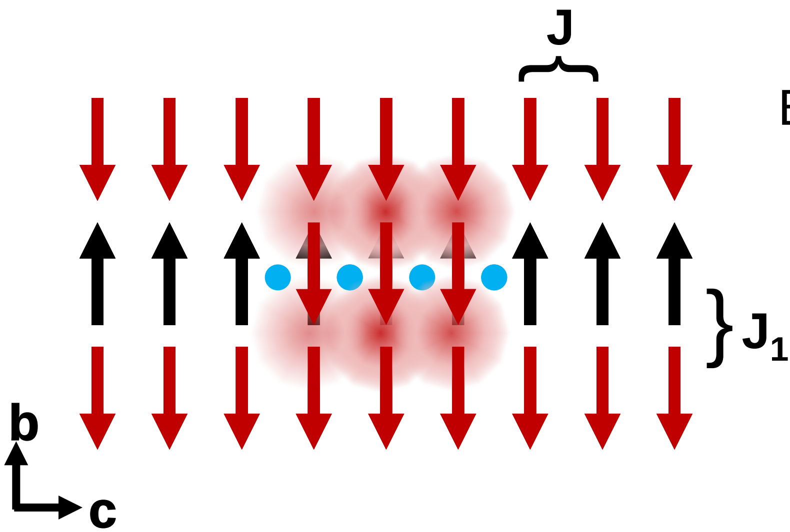
Spins point in a-c plane

- T_{N1} = 2.95 K, magnetic ordering, incommensurate
- T_{N2} = 1.97 K, antiferromagnetic ordering
- Interesting QCP @ H_b = 5.5 Tesla



T. Hanawa et. al., J. Phys. Soc. Jap. **63**, 2706 (1994)

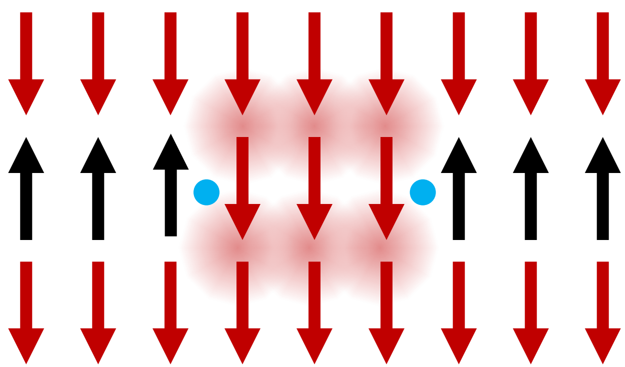
$CoNb_2O_6$ – Ising chain material - Excitations



Below T_{N2}, interchain mean field gives effective confining potential linear in separation of domain walls. Strong S_zS_{z+1} interaction.

$$\mathbf{E_n = 2(J/2) + n\, J_1}$$

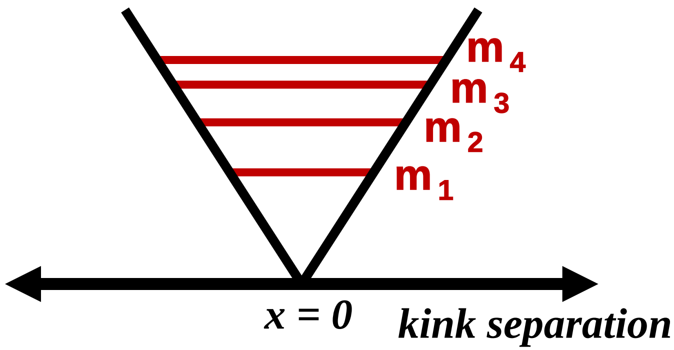
n is # of flipped spins



But real materials are not pure Ising! → S_x, S_y and inter-chain couplings give "kink" motion

$CoNb_2O_6$ – Ising chain material - Excitations

Rewrite equations of motion with kink as fundamental particle



$$V(x) = \lambda x$$

$$-\frac{\hbar^2}{\mu}\frac{d^2\varphi}{dx^2} + \lambda|x|\varphi = (m - 2m_0)\varphi \qquad (2)$$

$$m_j = 2m_0 + z_j\lambda^{2/3}\left(\frac{\hbar^2}{\mu}\right)^{1/3} \qquad j = 1, 2, 3, \ldots$$

Mapping to 1D Schrodinger equation with linearly confining potential

Solutions are Airy functions, z_j are the negative roots to Airy functions (z_j = 2.34, 4.09, 5.52, 6.79, 7.94, 9.02, …)

PHYSICAL REVIEW D VOLUME 18, NUMBER 4 15 AUGUST 1978

Two-dimensional Ising field theory in a magnetic field: Breakup of the cut in the two-point function

Barry M. McCoy
Institute for Theoretical Physics, State University of New York, Stony Brook, New York 11794

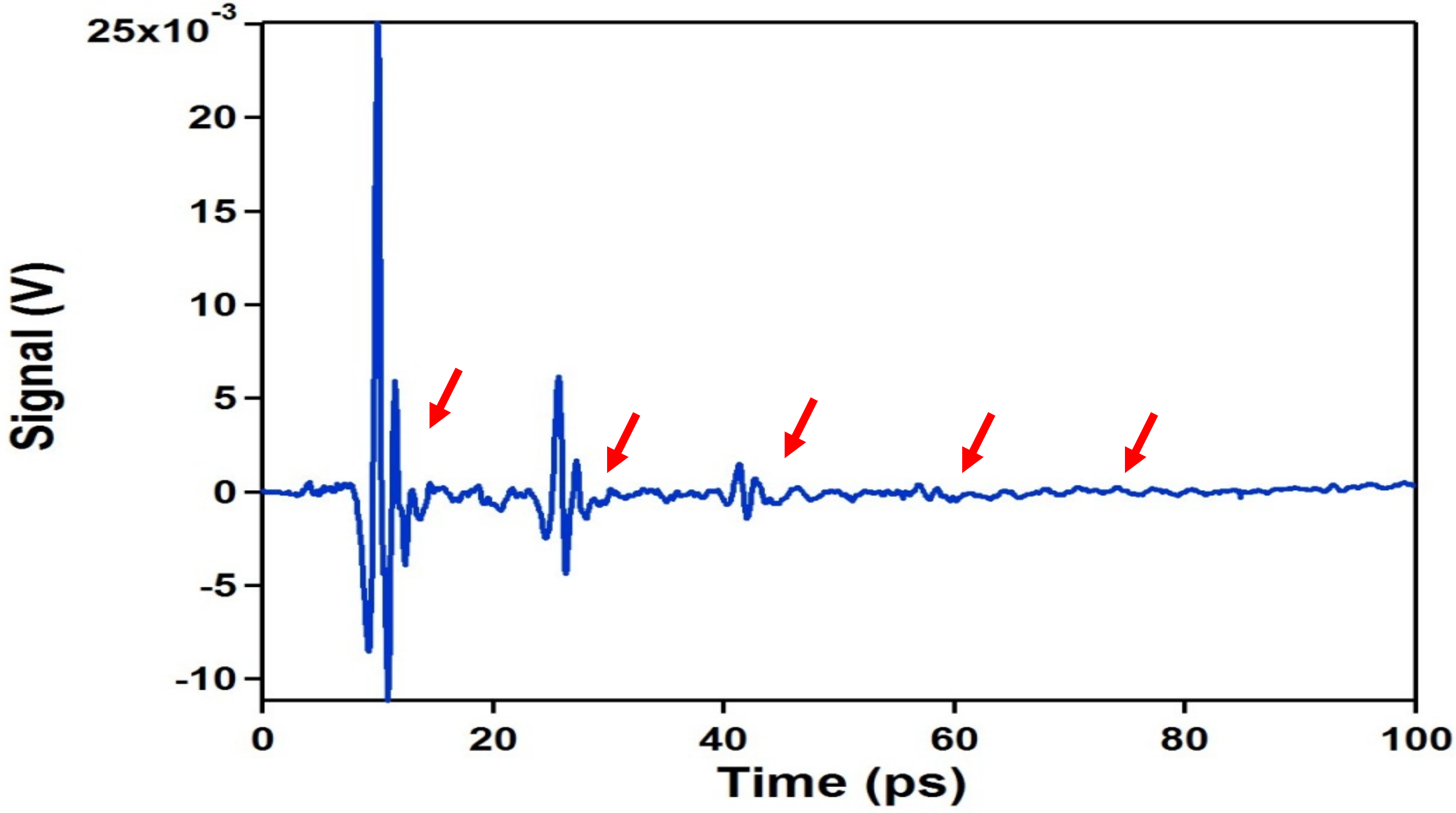
Tai Tsun Wu*
Institute for Theoretical Physics, Utrecht, The Netherlands

(Received 12 April 1978)

We demonstrate that the cut which is present as the leading singularity in the two-point function of the Ising field theory for $T < T_c$ and $H = 0$ breaks up into a sequence of poles for $H \neq 0$. Both the positions and the residues of the low-lying poles are calculated.

Analogy to quark confinement in QCD
kinks are quarks, bound state is a meson

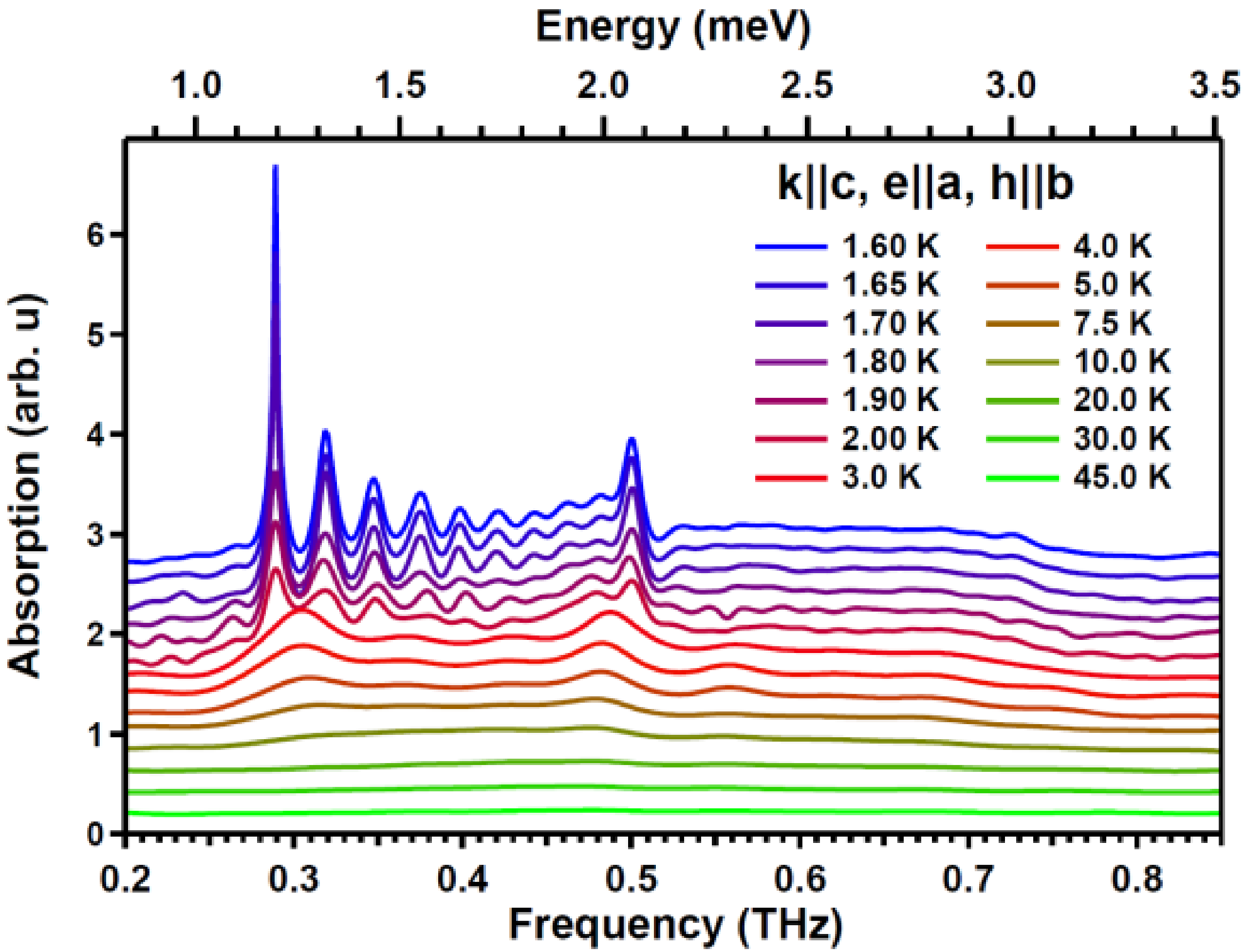
High resolution need → long scans



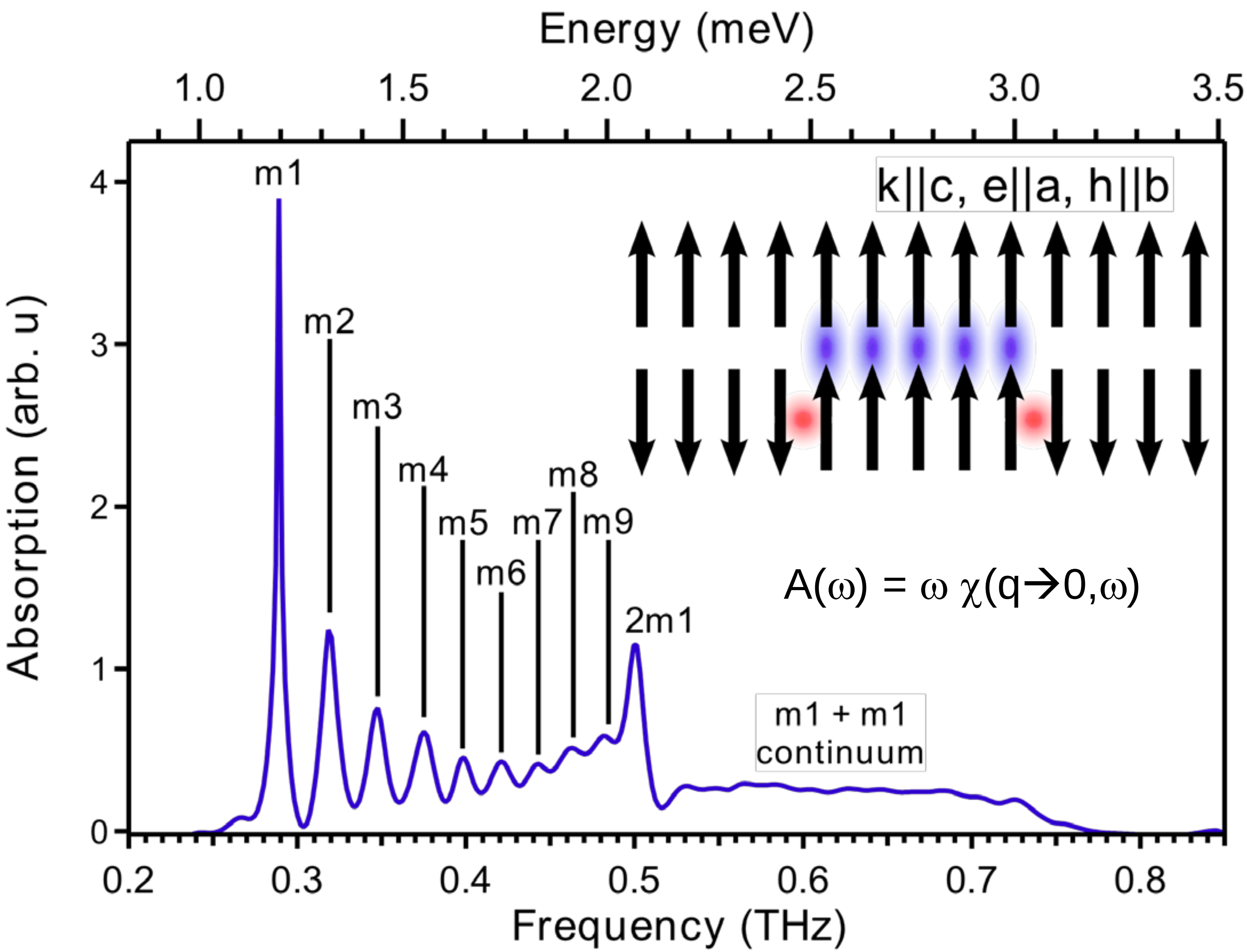
Time varying B-field from THz pulse can couple to spin → weak magnetic dipole absorption

$$\omega\, \chi(q \rightarrow 0,\omega) = -\ln(T(\omega))$$

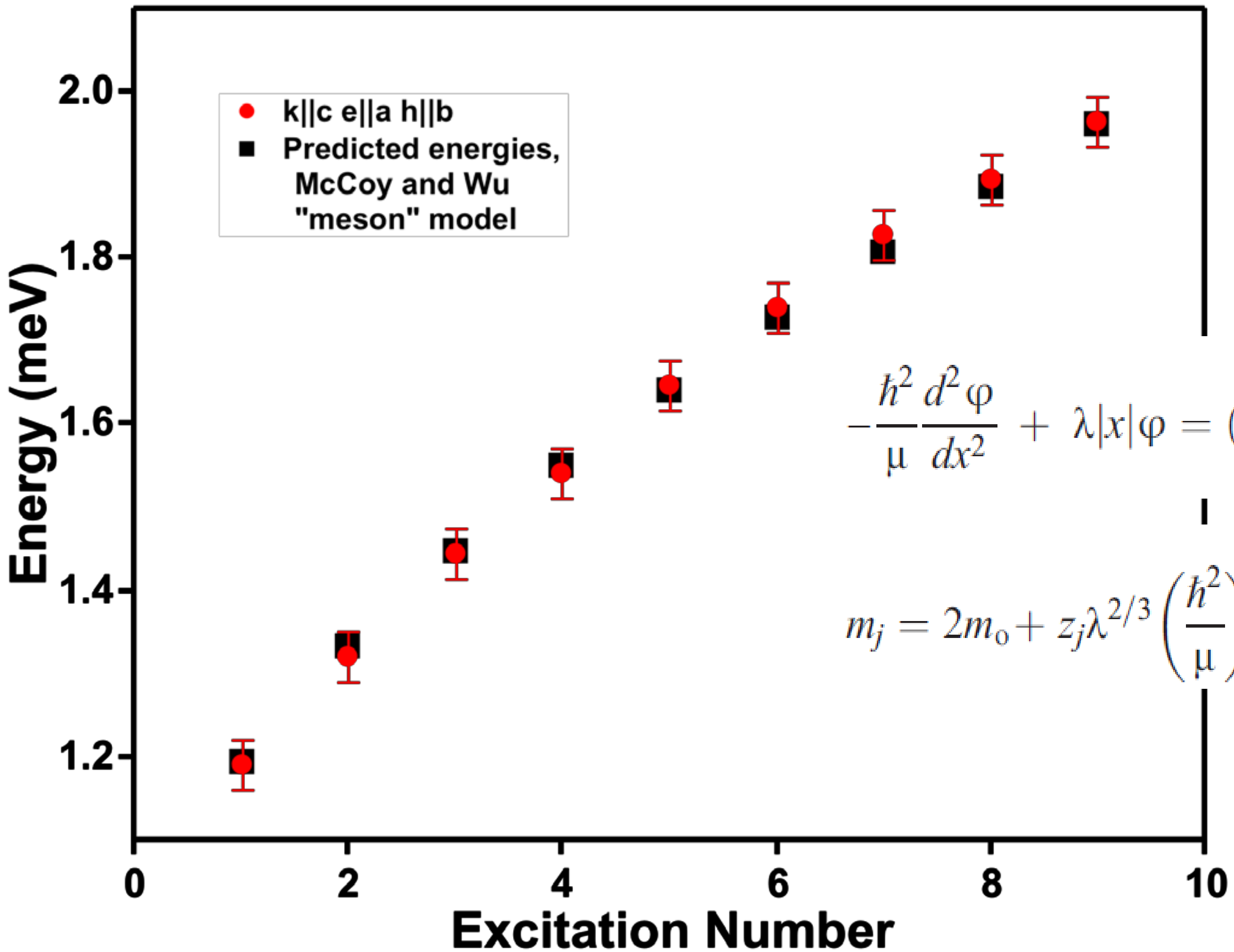
Temperature Dependence



Up to 9 “meson” bound states



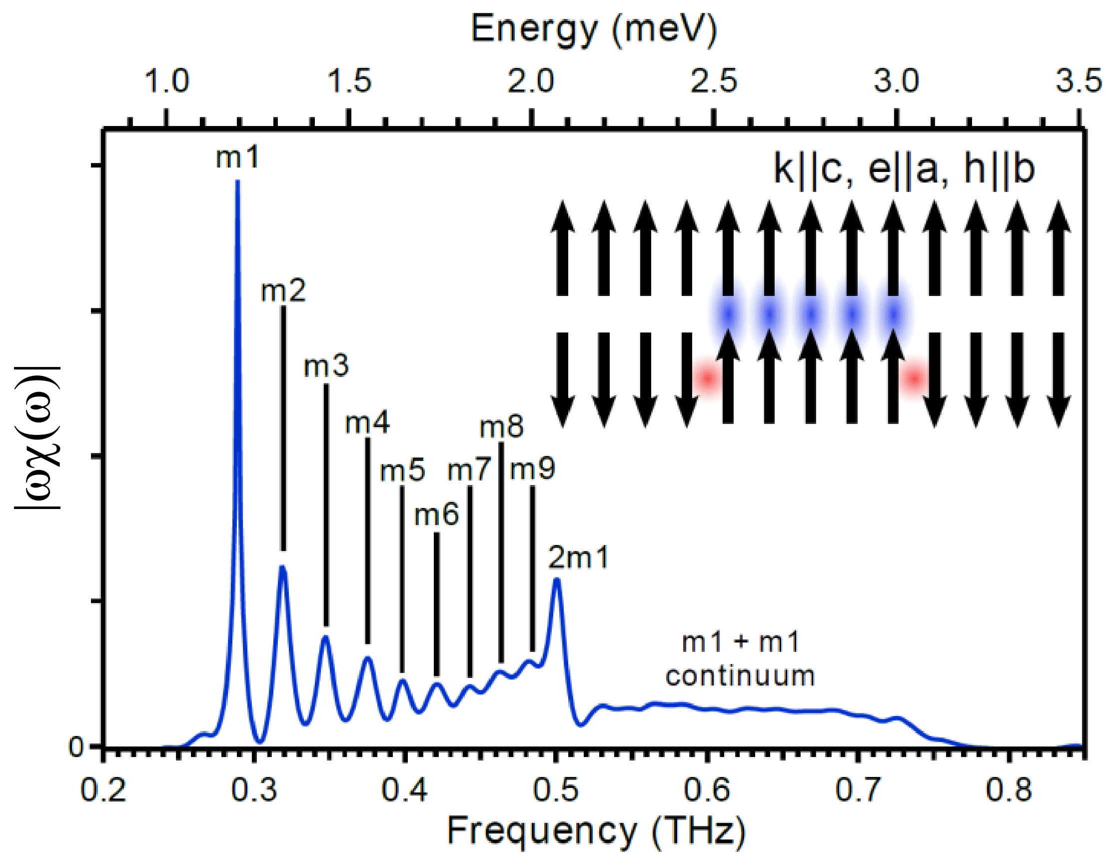
Comparison with McCoy and Wu "kink" model



$$-\frac{\hbar^2}{\mu}\frac{d^2\varphi}{dx^2} + \lambda|x|\varphi = (m - 2m_o)\varphi$$

$$m_j = 2m_o + z_j\lambda^{2/3}\left(\frac{\hbar^2}{\mu}\right)^{1/3} \qquad j = 1, 2, 3, \ldots$$

Beyond the Linear Chain

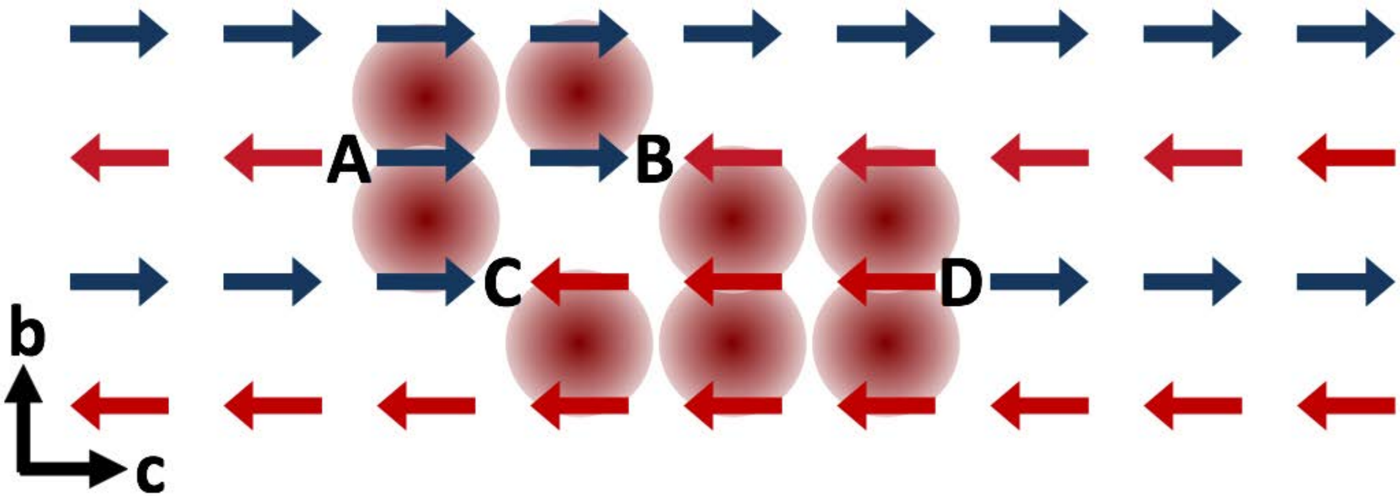


Quark analogy is useful!

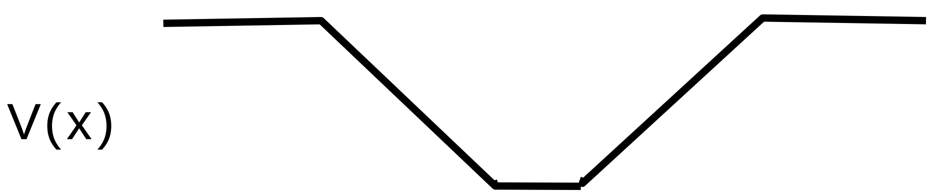
Hierarchy of meson states terminates at an energy twice the m1.
Confinement! No free quarks → m1+m1 continuum

Also “2m1” peak → 4 quark bound state.

Beyond the Linear Chain



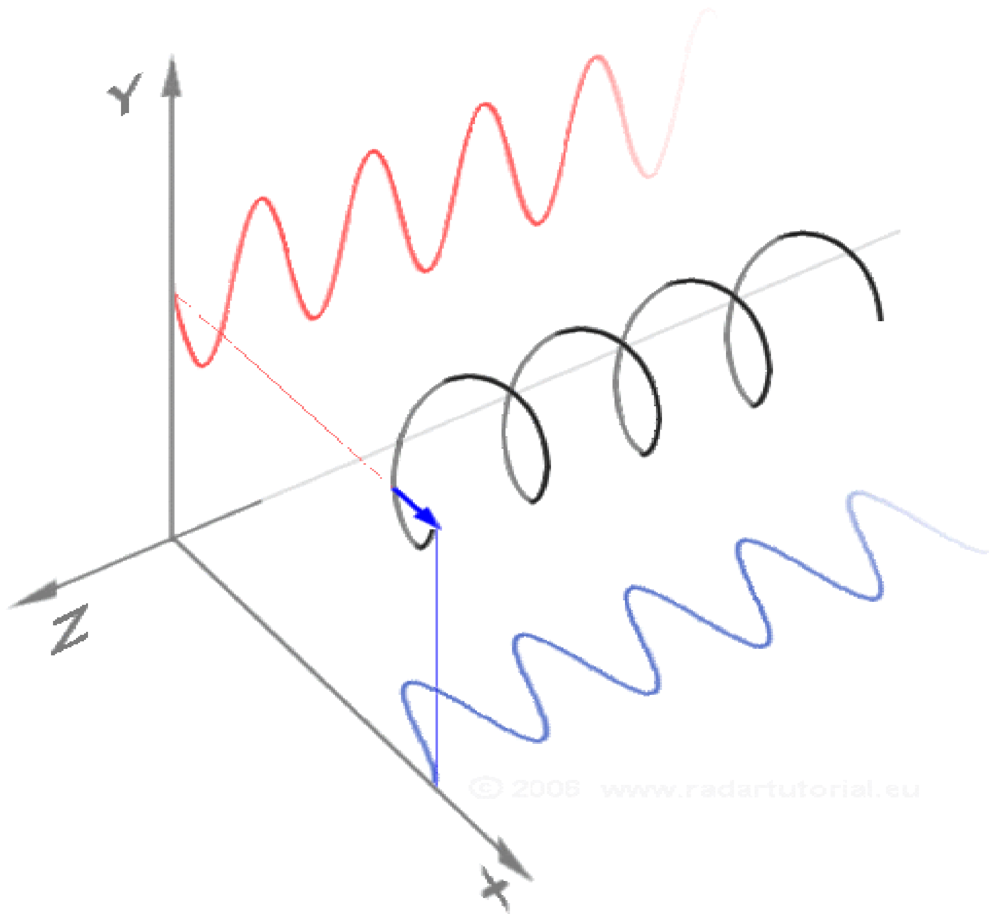
$$
\begin{aligned}
V(x_A, x_B, x_C, x_D) &= 4J_1[(x_D - x_C) + (x_B - x_A)] \;\; if \;\; x_D < x_A \;\; or \;\; x_B < x_C \\
&= 2J_1[|x_A - x_C| + |x_B - x_D|] + 2J_1[(x_D - x_C) + (x_B - x_A)] \;\; otherwise
\end{aligned}
$$



Ghosh and Tchernyshyov and calculation finds a “4 quark” bound state with large spectral weight

Example of attractive potential in 1D admits at least one solution

High Resolution THz Polarimetry

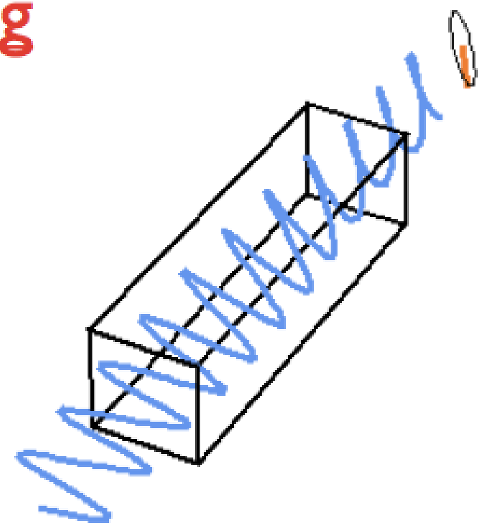


Polarization anisotropy is a probe of symmetry breaking

e.g. different response to different light polarizations

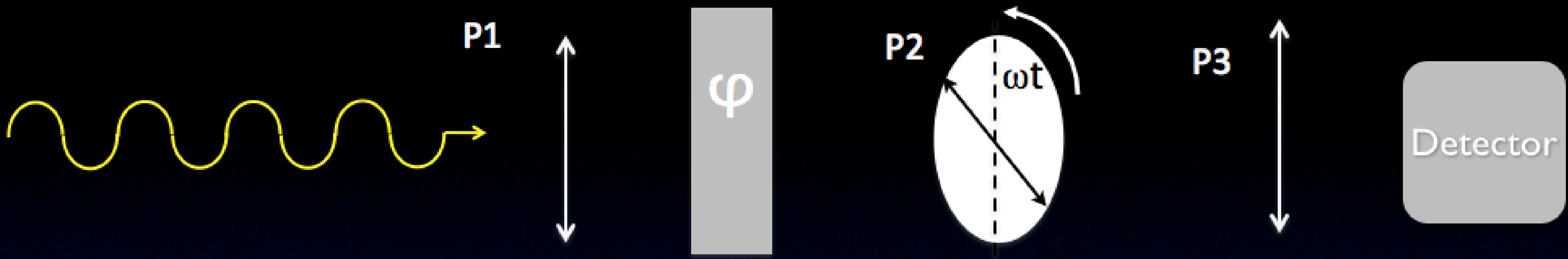
Implies a true static symmetry breaking

Scattering: Insensitive to fluctuations on scales that are long compared with the probing energy (h/ ΔE) and momentum resolution



Spectroscopy: gaps can result from partial order that is short range in both space and time.

Need more sensitive instrumentation!



Polarizer P2 spun at high speed (~10,000 rpm)
Lockin detection at 2ω → Allows phase sensitive detection of E_x and E_y

$|E_{12}| \sim |\cos(\omega t)|$ ← *Electric field going through P1 and P2*
$|E_{23} \sim |\cos(\omega t + \varphi)|$ ← *Electric field going through P2 and P3*

$$\begin{aligned}|E_{det}| &\sim |\cos(\omega t)\cos(\omega t + \varphi)| \\ &\sim \cos(\omega t)[\cos(\omega t)\cos(\varphi) - \sin(\omega t)\cos(\varphi)] \\ &\sim \cos^2(\omega t)\cos(\varphi) - \cos(\omega t)\sin(\omega t)\sin(\varphi) \\ &\text{with } \cos^2 a = \tfrac{1}{2} + \tfrac{1}{2}\cos 2a \text{ and } \cos a \cdot \sin a = \tfrac{1}{2}\sin 2a\end{aligned}$$ ← *Electric field at detector*

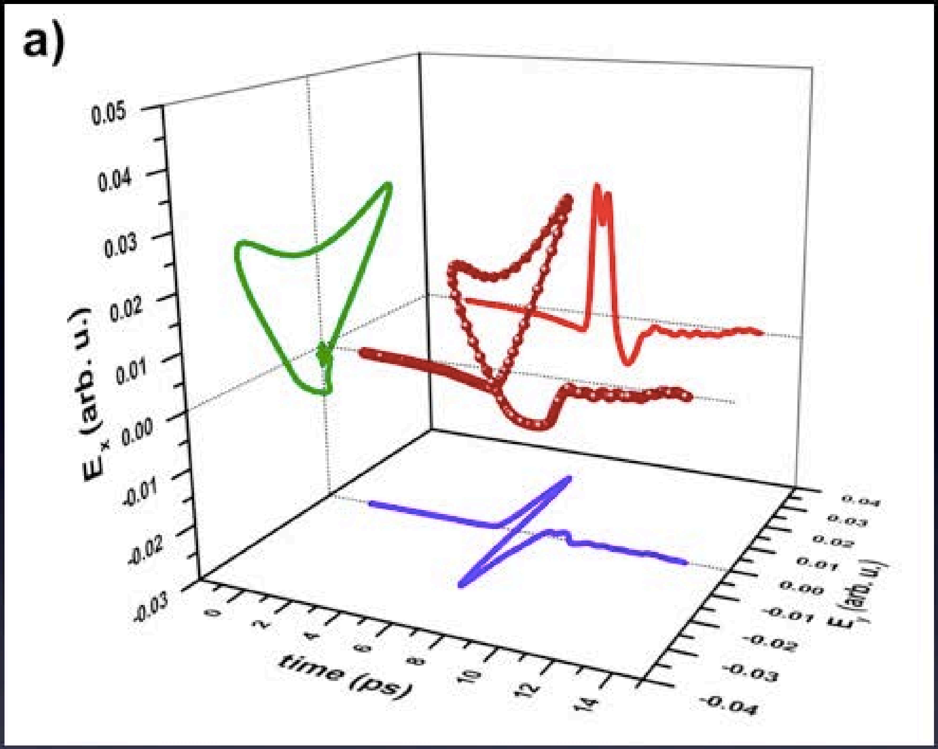
$$|E_{det}| \sim [\tfrac{1}{2} + \tfrac{1}{2}\cos(2\omega t)]\cos(\varphi) - [\tfrac{1}{2}\sin(2\omega t)]\sin(\varphi)$$

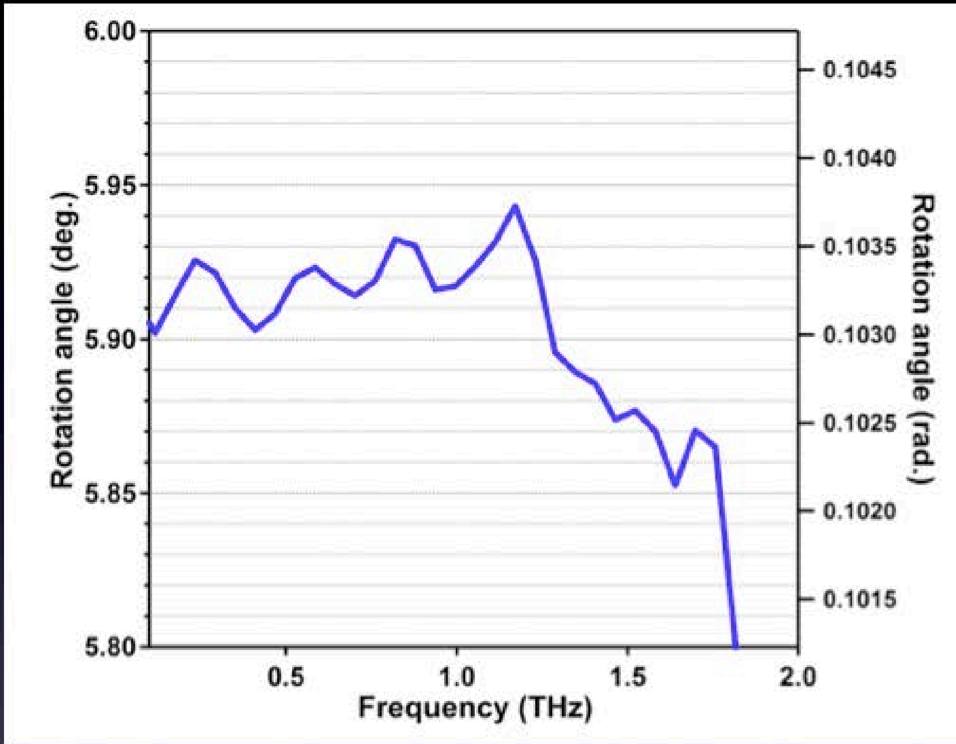
Lockin amplifier @ 2ω outputs in- and out-of-phase components at 2ω

$$R_{lockin} = A\int_0^{\tau} dt\ ([\tfrac{1}{2} + \tfrac{1}{2}\cos(2\omega t)]\cos(\varphi) - [\tfrac{1}{2}\sin(2\omega t)]\sin(\varphi)) \times (\cos(2\omega t) + i\sin(2\omega t))$$ ← *Complex quantity*

In- and out-of-phase components in lockin are proportional to E_x and E_y

High precision THz polarimetry capabilities



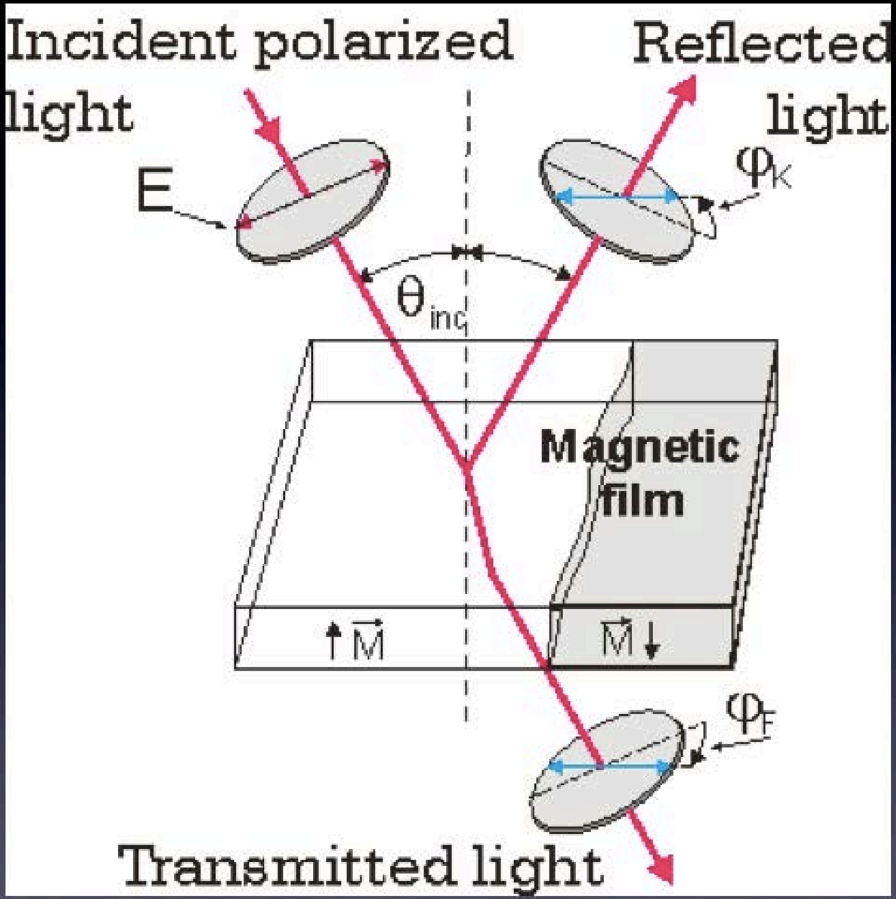


Arbitrary elliptical polarization introduced by R-cut sapphire

Example: polarizer used as "sample" ~20 mdeg resolution

(I think we can still do an order magnitude better precision... stay tuned)

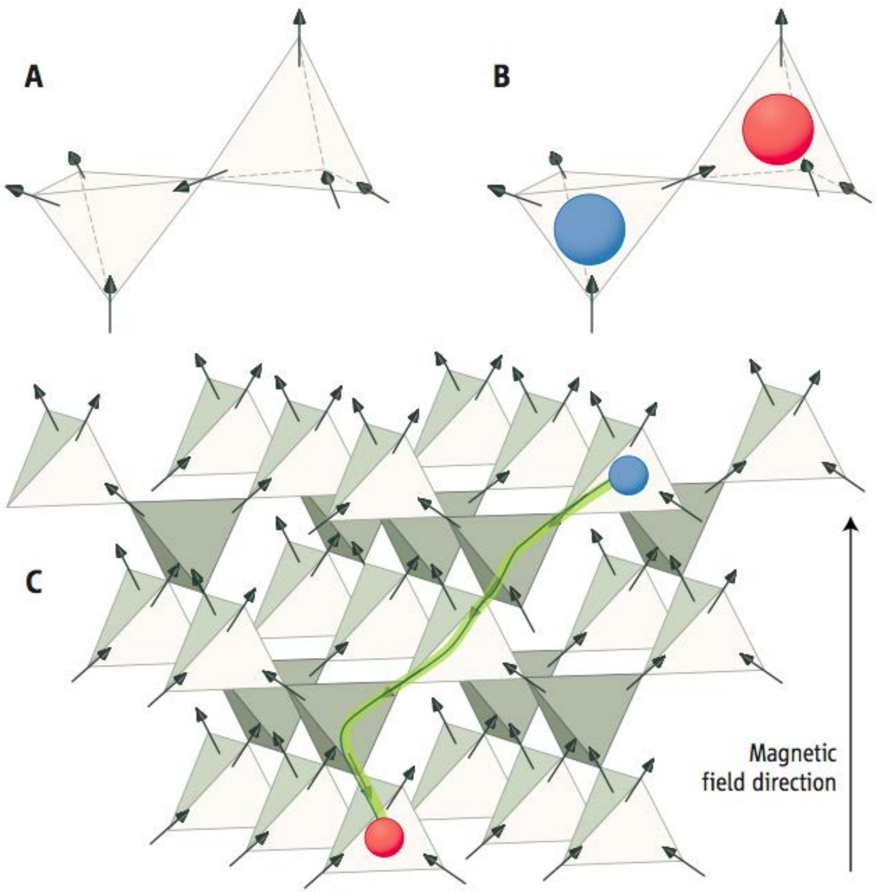
Rotation of Polarization of Transmitted and Reflected Light



Kerr effect
Rotation Reflection
John Kerr (1870-ish)

Faraday effect
Transmission Reflection
Michael Faraday (1845)

Magnetic monopoles in spin-ice



Ex. $Ho_2Ti_2O_7$, $Dy_2Ti_2O_7$, $Yb_2Ti_2O_7$

2 in – 2 out is the vacuum state – “ice-rule” like water ice with protons

Flipped spin is dipole excitation.

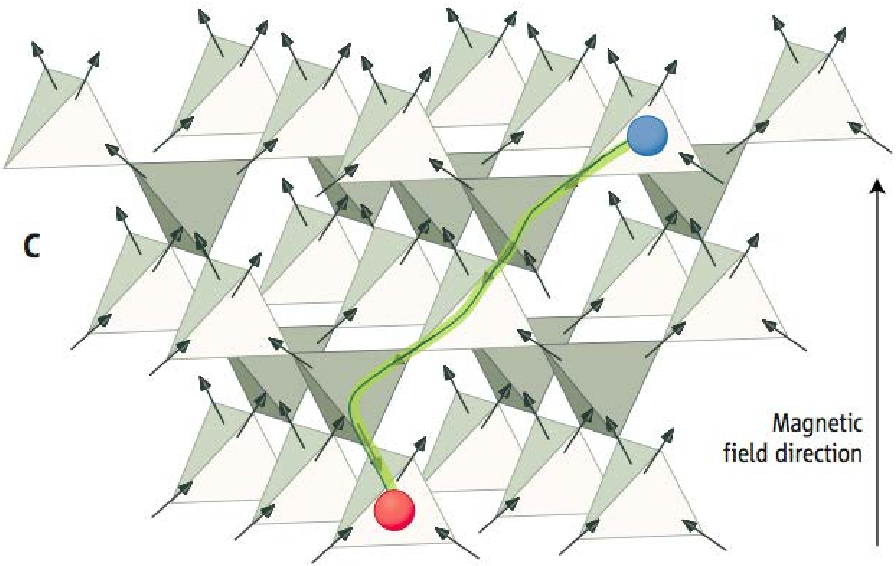
Can separate ends of dipole with emergent Coulomb confinement

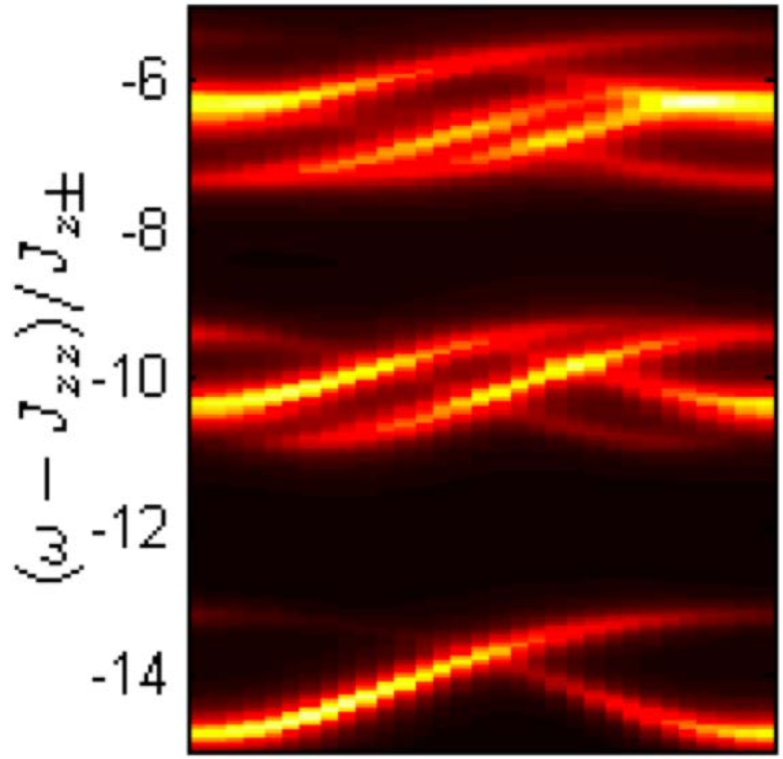
Ryzhkin (2005); Castelnovo, Moessner, and Sondhi(2008)

$$V(r_{\alpha\beta}) = \begin{cases} \frac{\mu_0}{4\pi}\frac{Q_\alpha Q_\beta}{r_{\alpha\beta}} & \alpha \neq \beta \\ \frac{1}{2}\upsilon_o Q_\alpha^2 & \alpha = \beta \end{cases}$$

Novel excitations in quantum spin ice

In “quantum” spin ice, small spins and interactions beyond Ising allow motion of dipoles





Wan and Tchernyshyov
PRL 2012

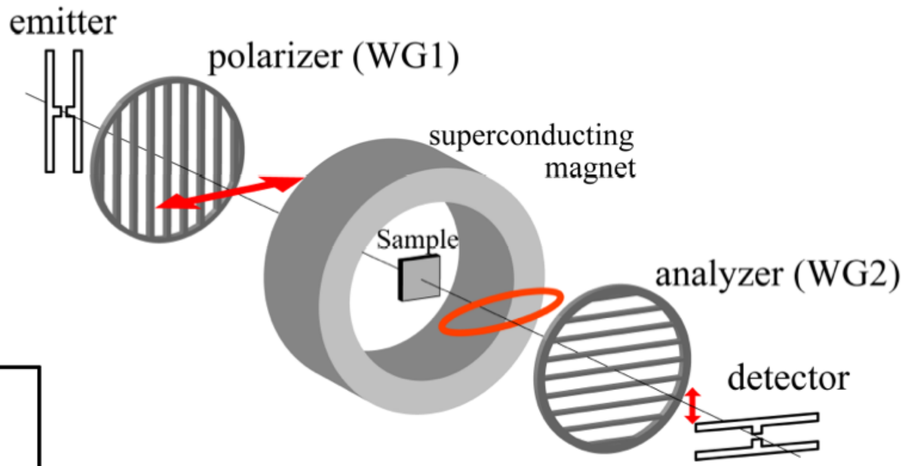
Applied magnetic field gives a tension to strings connecting dipoles

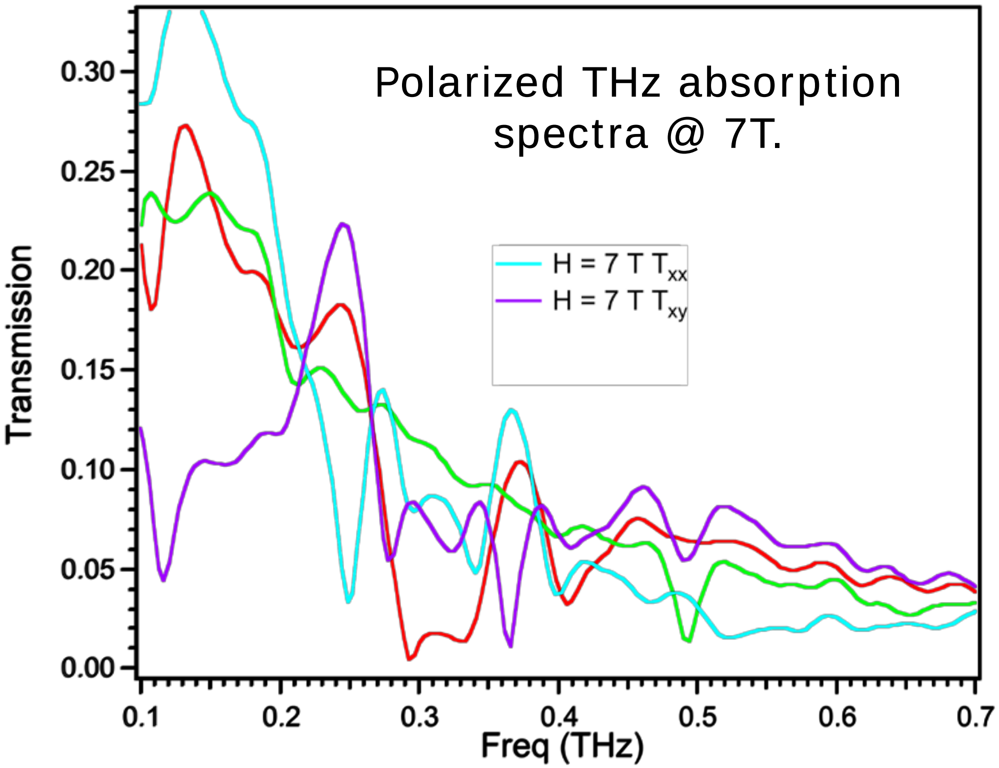
Novel extended object excitations

Novel excitations in quantum spin ice

$$\begin{bmatrix} T_{xx} & T_{xy} \\ T_{yx} & T_{yy} \end{bmatrix} \begin{bmatrix} E_x^i \\ E_y^i \end{bmatrix} = \begin{bmatrix} E_x^r \\ E_y^r \end{bmatrix}$$

We can measure all components of T matrix



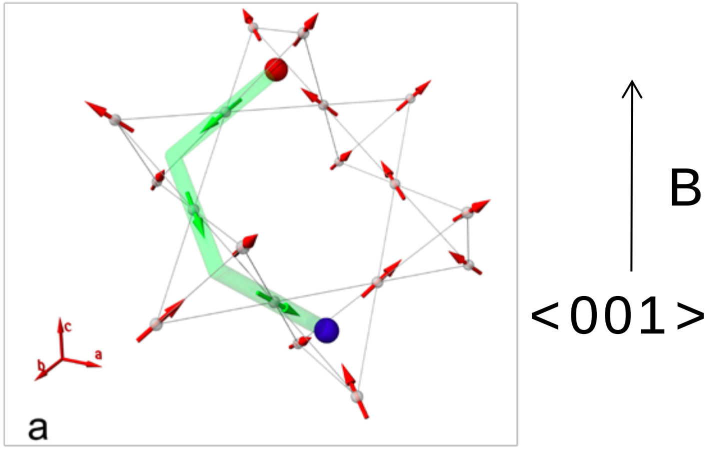


Complex phase information from TDTS allows decomposition into circular eigenstates →

$$T_R = \frac{T_{xx} + iT_{xy}}{\sqrt{2}}$$

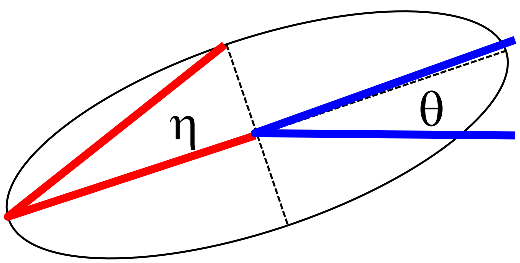
$$T_L = \frac{T_{xx} - iT_{xy}}{\sqrt{2}}$$

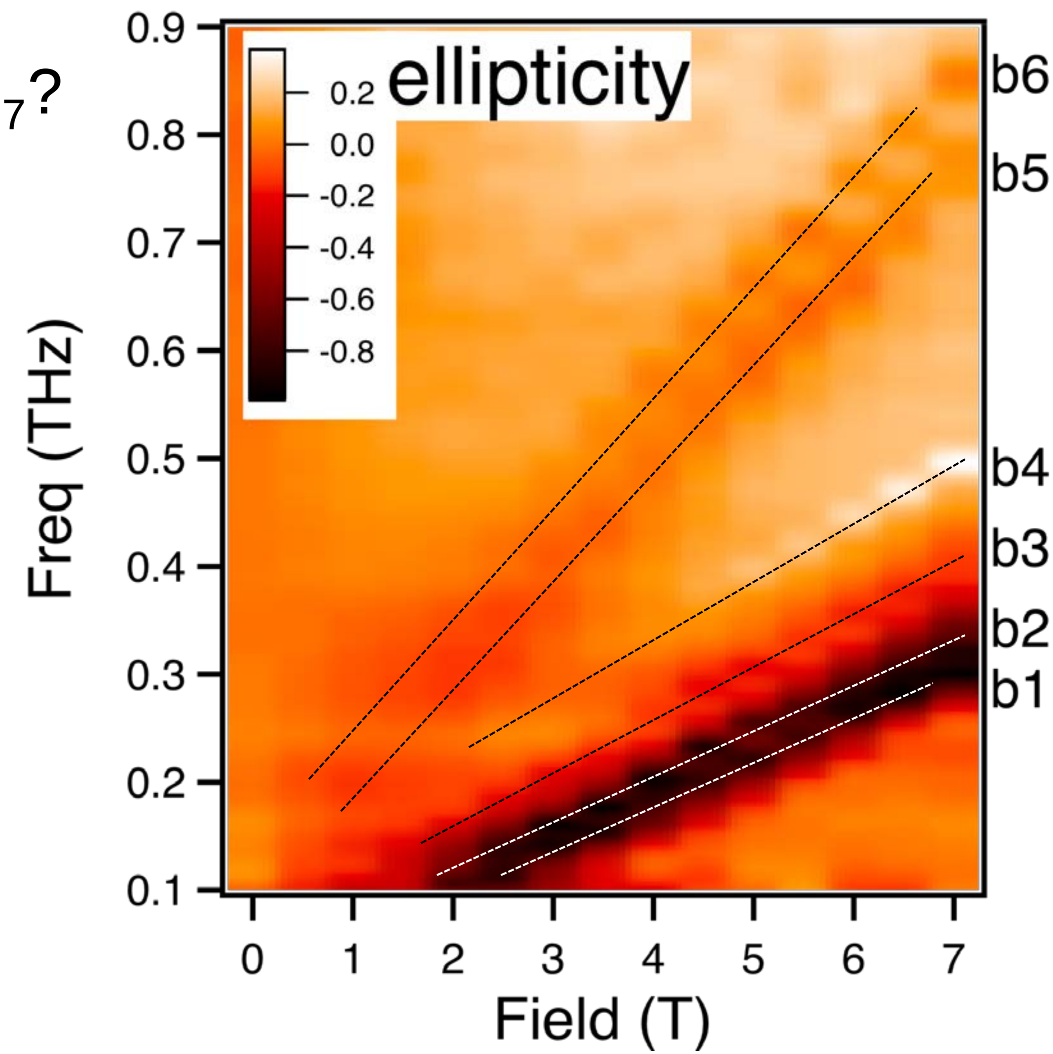
String excitations in quantum spin ice $Yb_2Ti_2O_7$?



Useful quantity is the ellipticity.

$$\eta = \frac{t_r - t_l}{t_r + t_l}$$

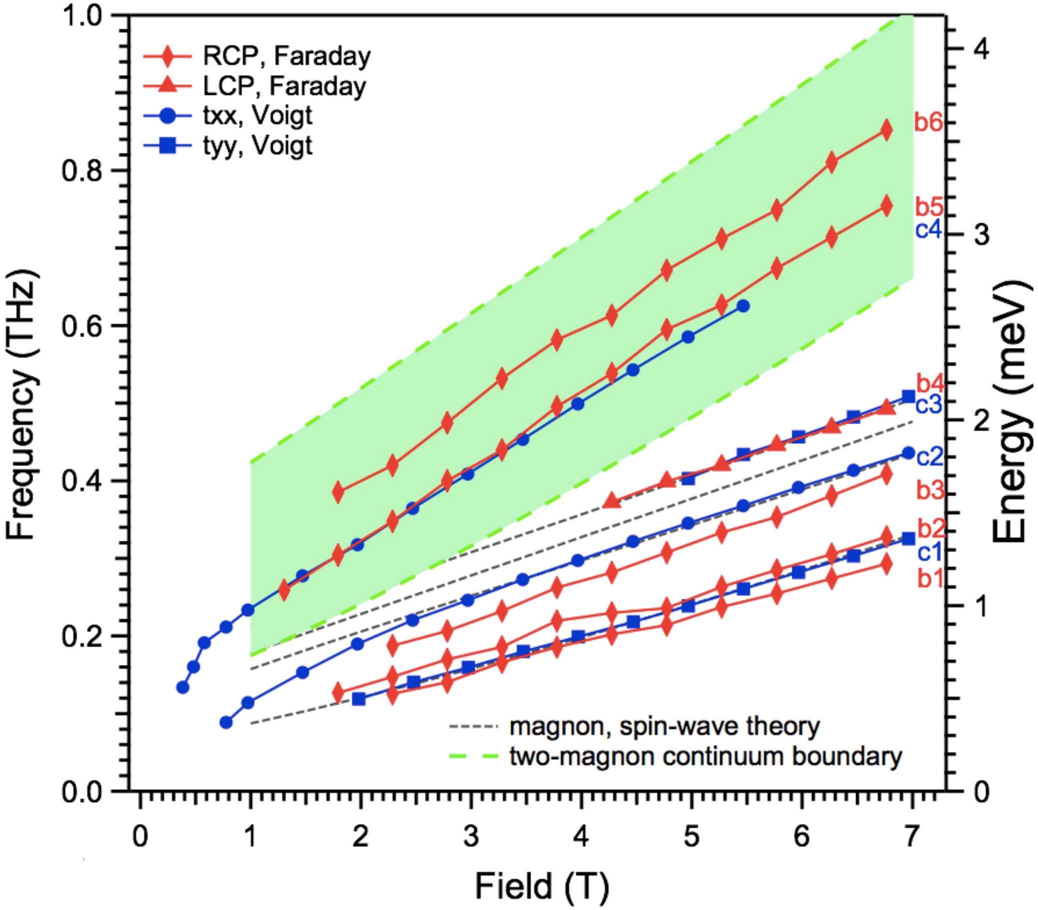


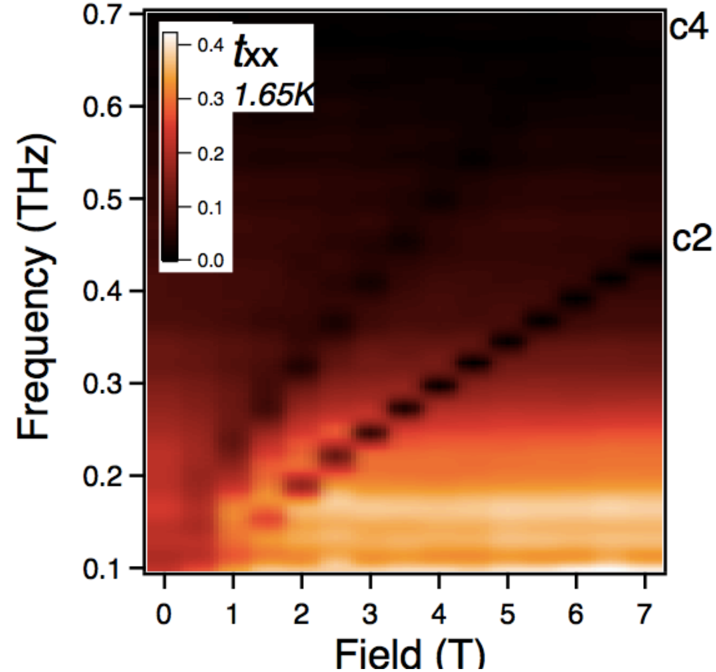


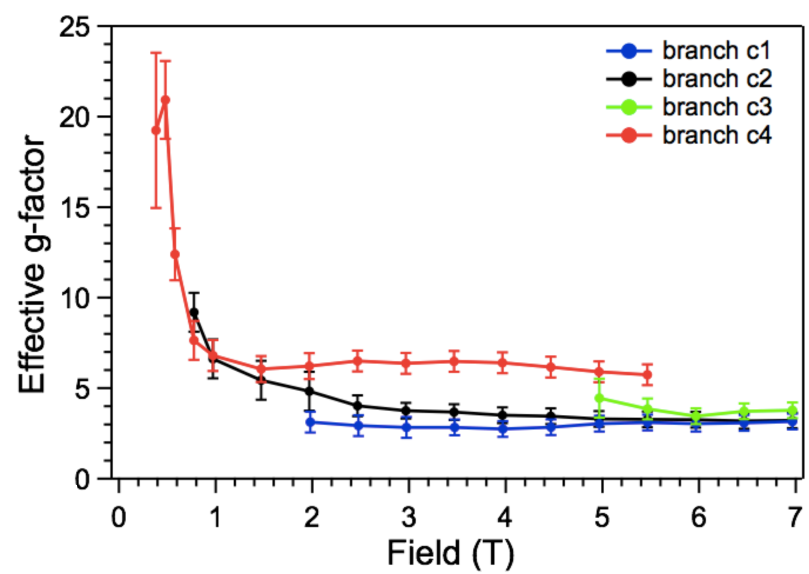
One expects max 4 excitations (2 only from spin wave calculations)
We see 6 and measure their polarizations.

Ratio of g-factors show higher multiplet is strings of length 2.

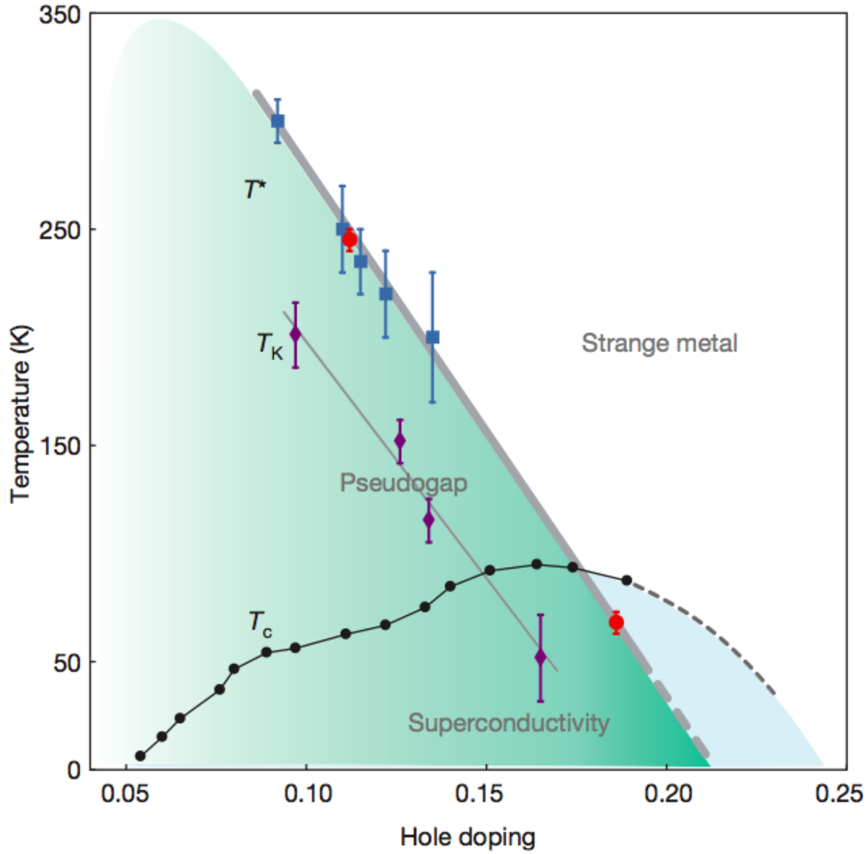
String excitations in quantum spin ice $Yb_2Ti_2O_7$?



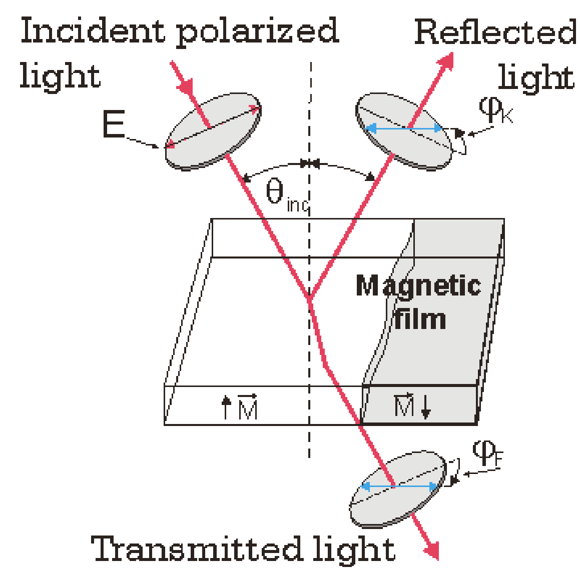




Origin of pseudogap in underdoped cuprates?



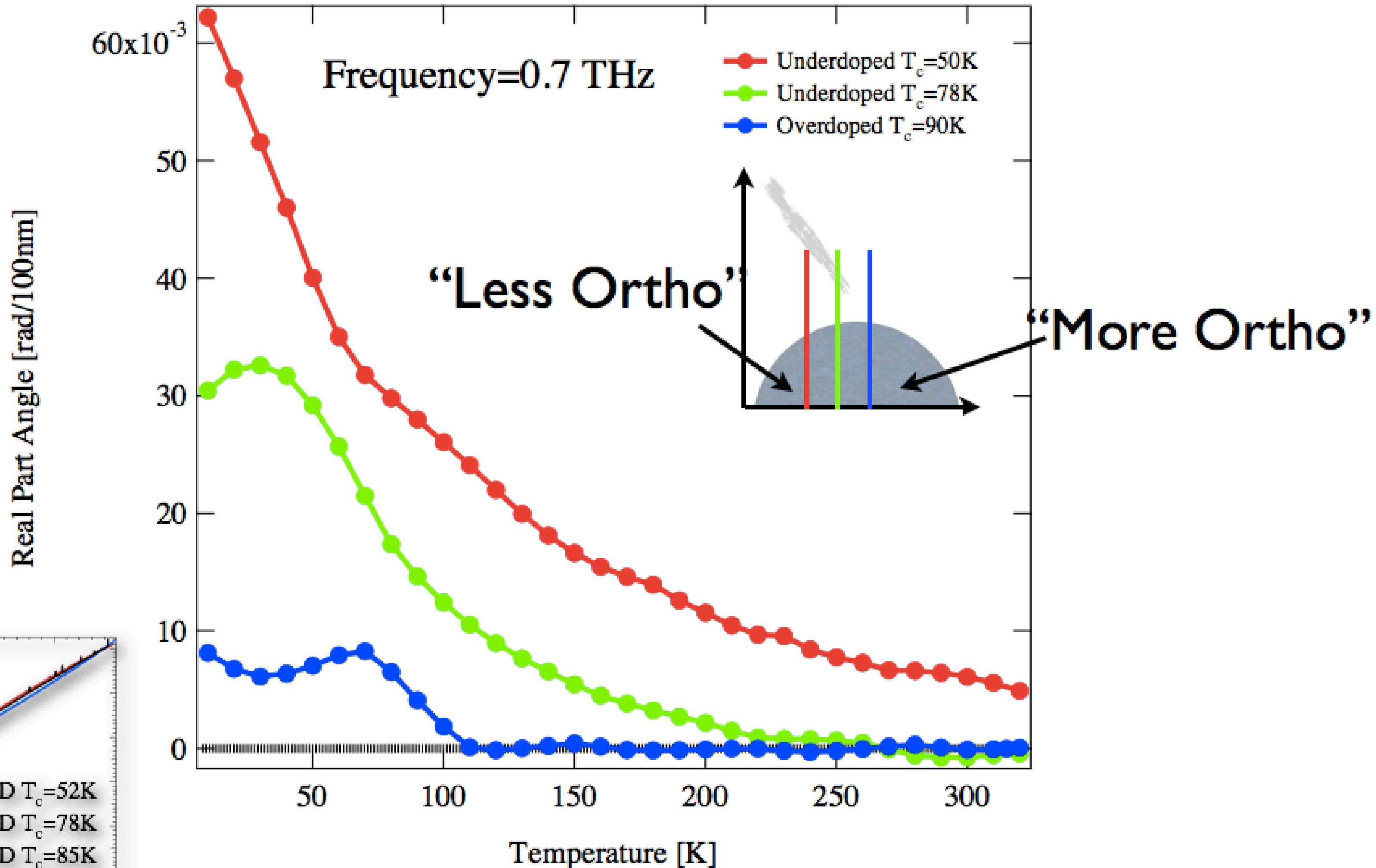
Example $La_{2-x}Sr_xCuO_4$

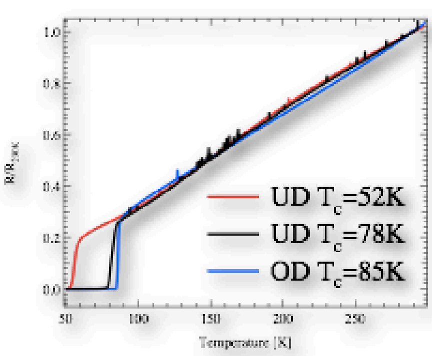


THz has contributed greatly to pseudogap discussion. Ruled out anomalous high-temperature superconducting fluctuations (Bilbro thesis 2012)

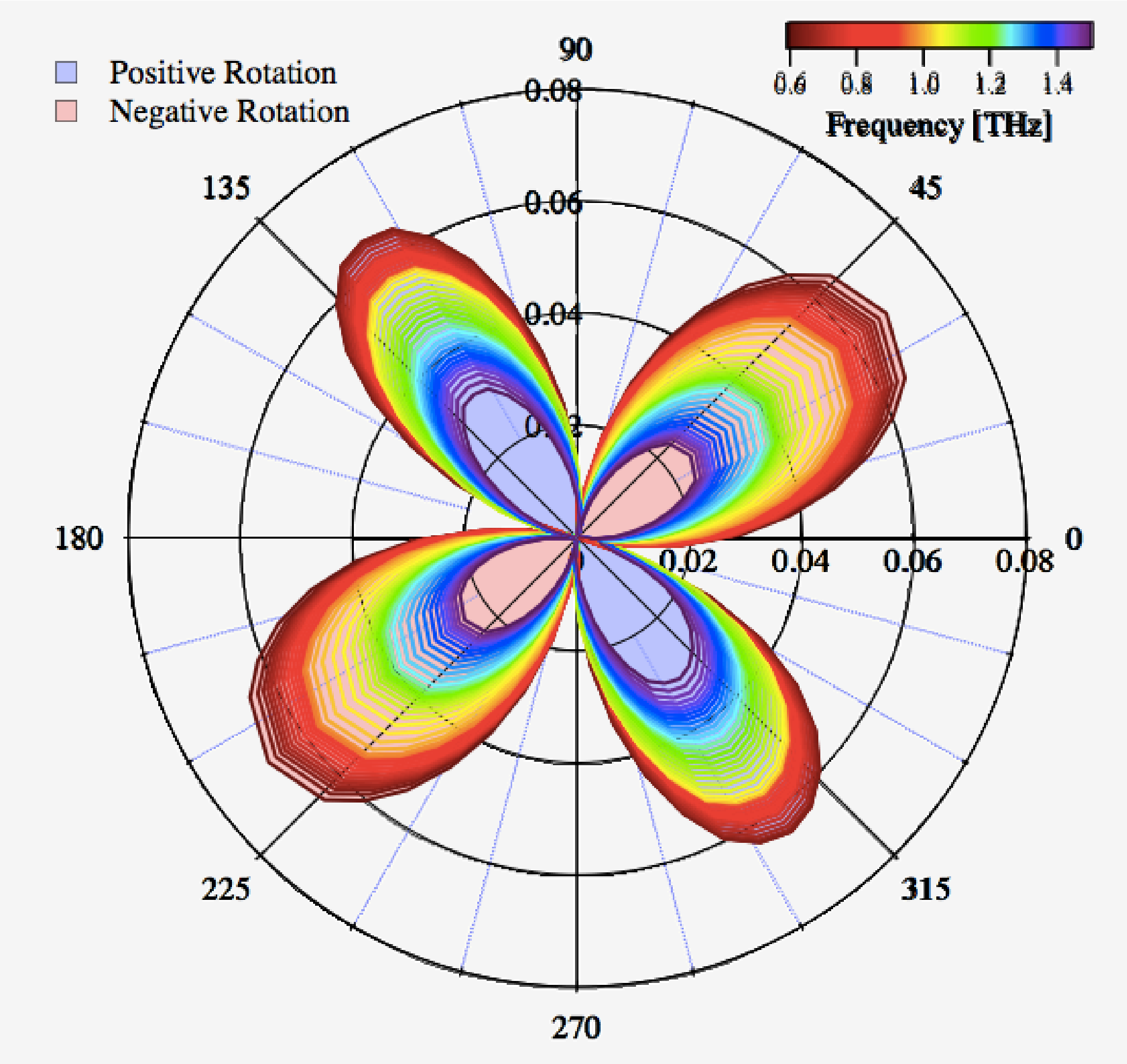
Polarization anisotropies are signature of true long range symmetry breakings.

Doping Dependence

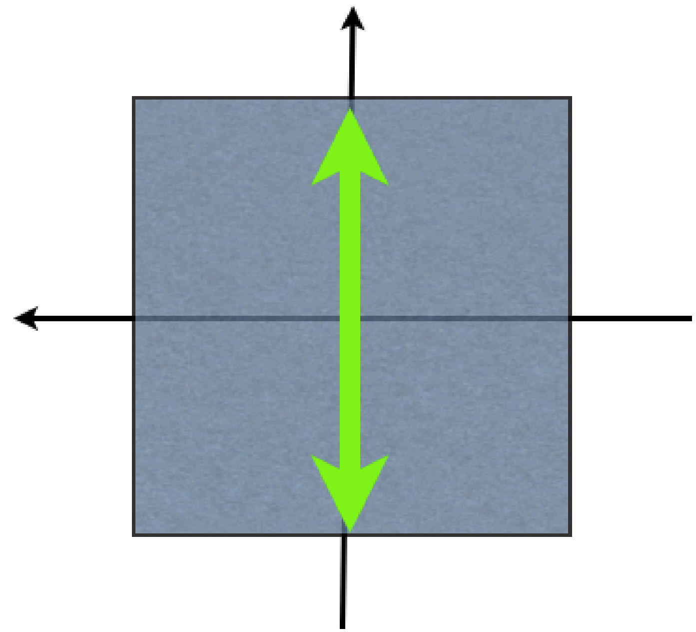




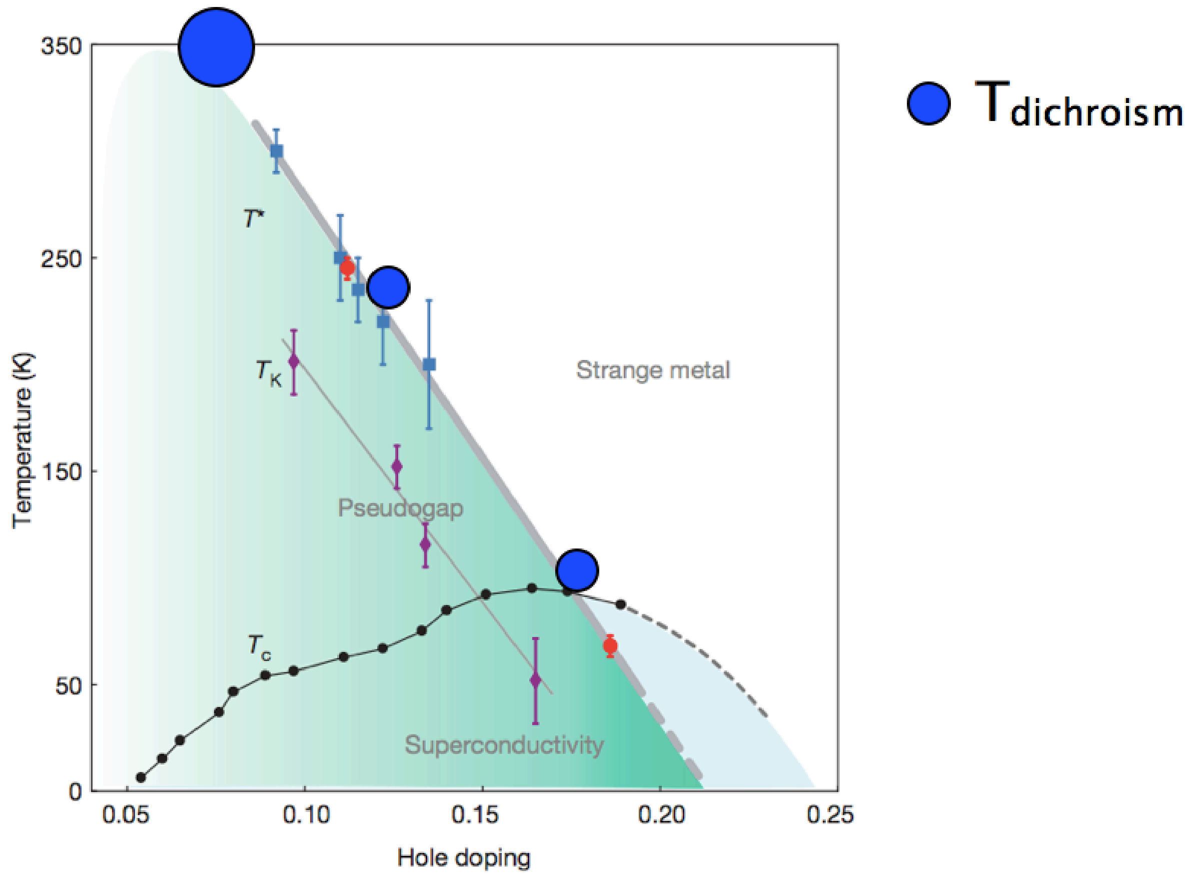
T=10K



Positive Rotation
Negative Rotation
Frequency [THz]
0.6
0.8
1.0
1.2
1.4
90
135
45
180
0
225
315
270
0.08
0.06
0.04
0.02
0
0.02
0.04
0.06
0.08



Phase Diagram



Shekhter et al. Nature 2013

1D oriented electronic nematic phase? e.g. stripes

Magnetoelectric effects? Orbital currents?

All papers available on arXiv

Electrodynamics of correlated electron systems:
Based on 2008 Boulder School for Condensed Matter and Materials Physics Lectures

N. P. Armitage

Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, MD 21218, USA

Contents

incredibly broad range means that a blizzard of experimental techniques and analysis methods are required for the characterization of correlated systems with optical techniques.

This short review and lecture notes attempt to lay out a brief summary of the formalism, techniques, and analysis used for 'optical' spectroscopies of correlated electron systems. They are idiosyncratic, occasionally opinionated, and - considering the breadth of the subject - incredibly brief. Unfortunately, there is no single com-

_Available at http://arxiv.org/abs/0908.1126

PHYSICAL REVIEW B **90**, 035135 (2014)

Constraints on Jones transmission matrices from time-reversal invariance and discrete spatial symmetries

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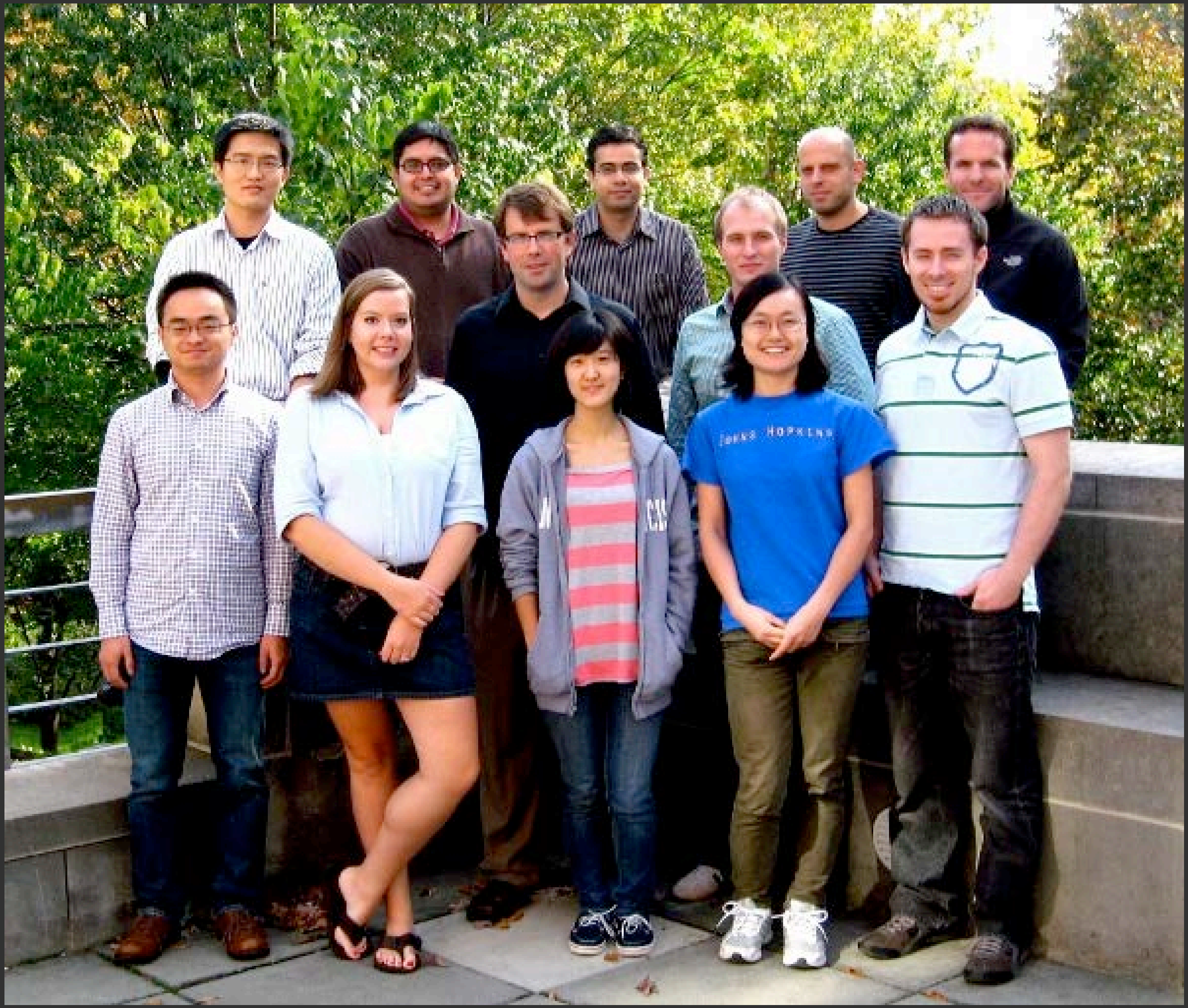
(Received 26 November 2013; published 28 July 2014)

Acknowledgements



Also … Collin Broholm, Tyrel McQueen, Oleg Tchernyshyov, Zlatko Tesanovic, Seyeed Koopayeh, Anirban Ghosh, Se Kwon Kim, John Sheckelton, Jamie Neilson, Bob Cava

And …Ivan Bozovic (BNL), Sambandamurthy Ganapathy (UB), John Cerne (UB), Andrea Markelz (UB), Seongshik Oh (Rutgers)

Conclusions…

-Discussed our evidence for a topological phase transition in topological insulators

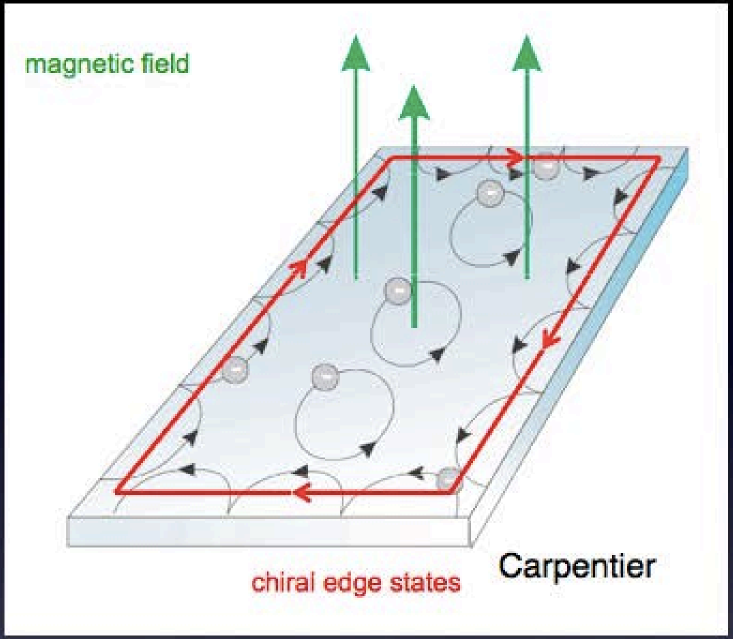
-Demonstrated novel “kink” excitations in linear spin-chains

-Evidence string excitations in spin ice?

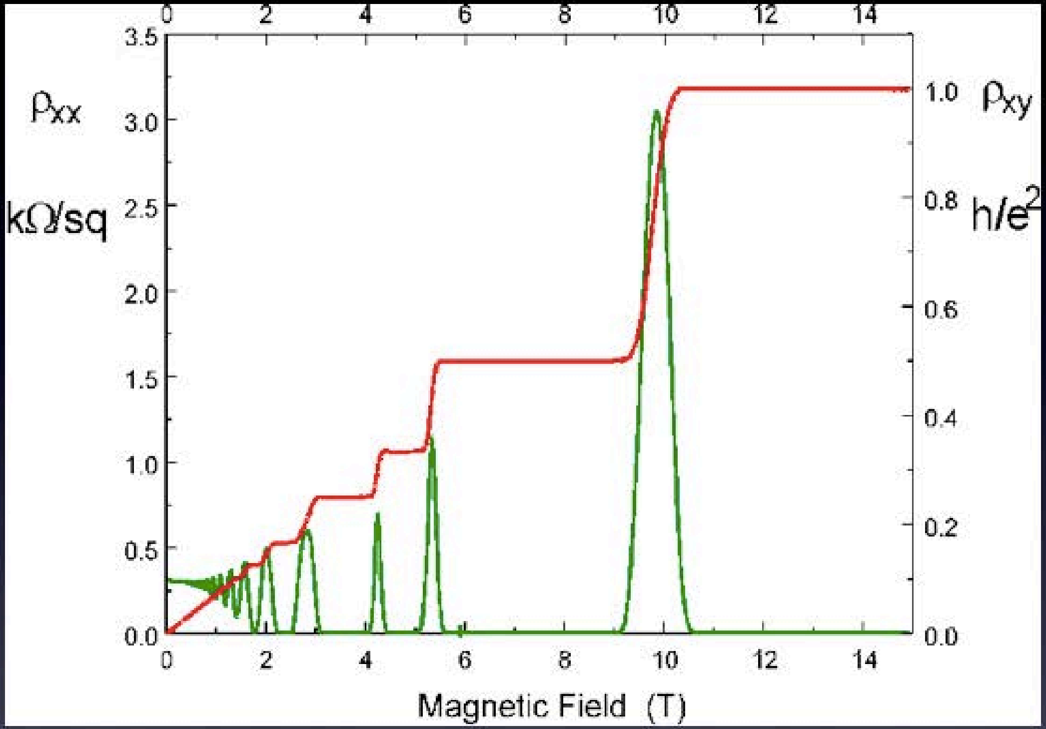
-Presented evidence for anisotropic conductivity in cuprate superconductors (nematicity? mangetoelectricity?)

…all the while emphasizing the role that new technology plays in our ability to make measurements

The Quantum Hall effect showed states can be different, despite same symmetry



2D Cyclotron Motion



$$\rho_{xy} = \frac{1}{n}\frac{h}{e^2}$$

- Different plateaus are different states with the **same** symmetry,
- Quantization exact to $1/10^9$ → Cannot be same state
- Wavefunctions have different topology - net curvature of wavefunction in wavefunction space
- 1st discovered topological insulator

New instrumentation directions for THz

Available online at www.sciencedirect.com
ScienceDirect
Optics Communications 281 (2008) 527–532
ELSEVIER
Optics Communications
www.elsevier.com/locate/optcom

Terahertz imaging for non-destructive evaluation of mural paintings

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[c] Center for Research and Restoration, The Louvre Museum, Paris, France
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Received 3 July 2007; received in revised form 4 October 2007; accepted 11 October 2007

Pigment under 5mm plaster

Example: Sanguine has very weak MIR signatures, but strong THz ones