

Possibilities for 2D coherent THz spectroscopy in quantum materials: electron glasses, spin liquids and *superconductors*

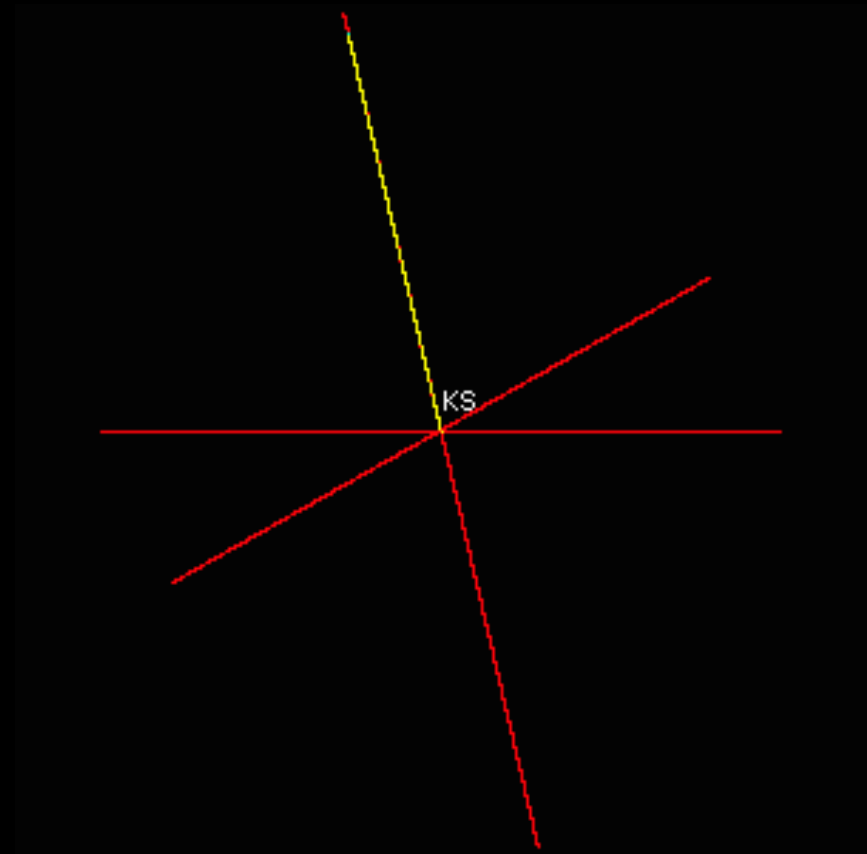
N. Peter Armitage
The Johns Hopkins University

We can measure many of their correlations of quantum materials only imperfectly with existing tools. A promising direction is *nonlinear* response e.g.

$$P_i = \chi_{ijk}^e E_j$$

$$P_i = \chi_{ijkl}^e E_j E_k E_l$$

$$M_i = \chi_{ijkl}^m B_j B_k B_l$$





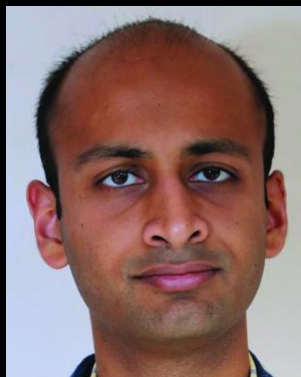
Fahad Mahmood
(JHU → UIUC)



Dipanjan Chaudhuri
(JHU)



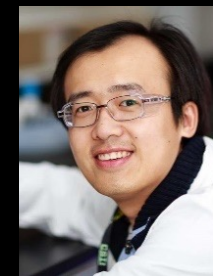
David Barbalas
(JHU)



Rahul Nandkishore
(CU Boulder)



Ivan Bozovic (BNL)



Xi He (BNL)

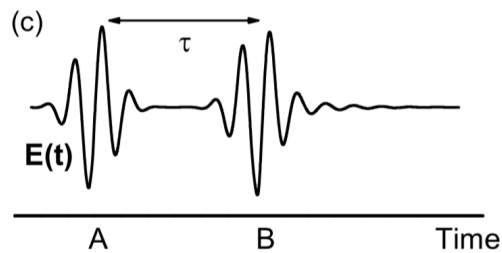


Conclusions

- 2D coherent THz spectroscopy is a new nonlinear $\chi^{(3)}$ technique with vast potential for quantum materials
- Can get unique information about spin liquids (experiments ongoing... theory now only)
- Photon echo observed in a electron glass $\rightarrow$ not continuable to weakly interacting disordered “Anderson insulator”; a “non-Fermi glass”
- Cuprate superconductors show a large regime of normal state $\chi^{(3)}$ nonlinearity; anomalous response unlike that of normal metals

2D Coherent Terahertz Spectroscopy

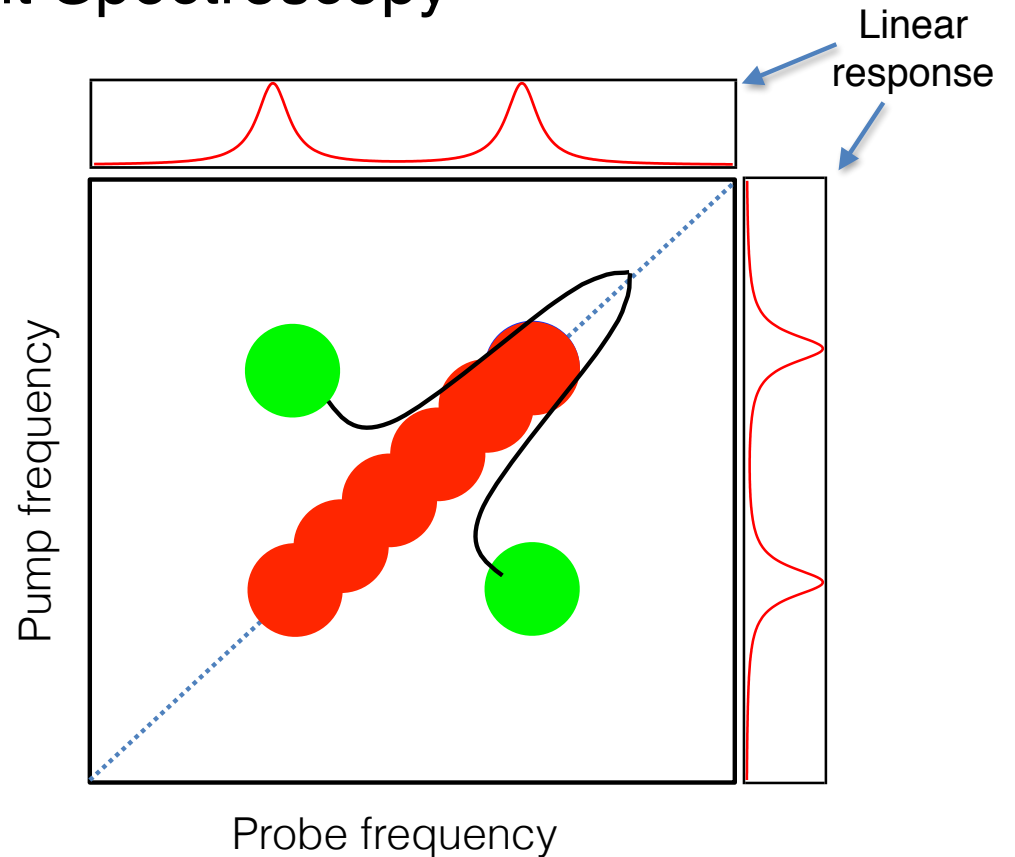
2D Coherent Spectroscopy



$$E_{NL} = E_{AB} - E_A - E_B$$

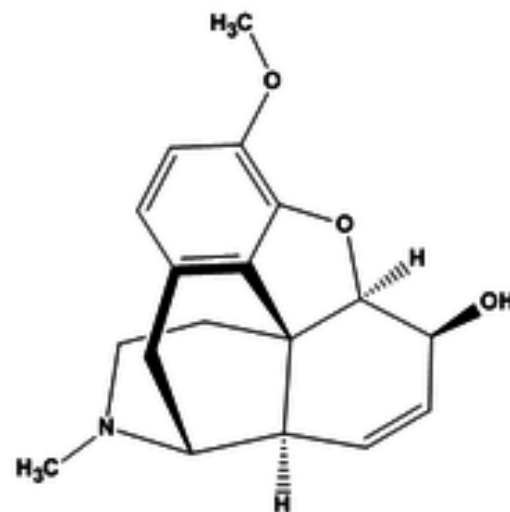
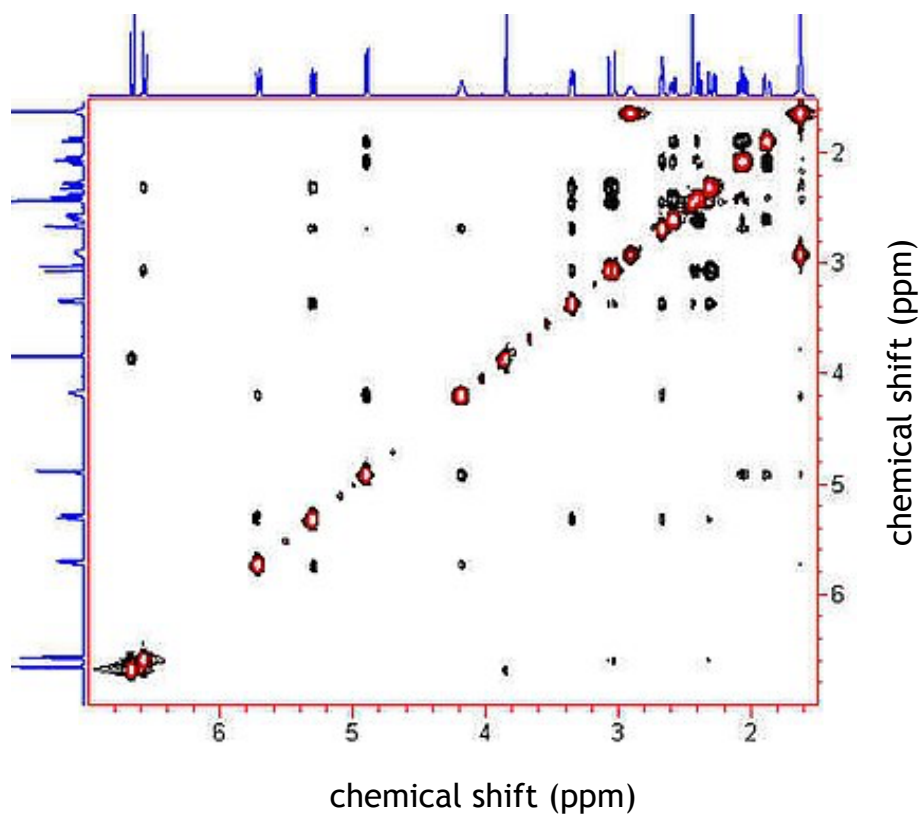
Measure nonlinear response and plot
as function of pump and probe
frequencies

Anti-diagonal width can be intrinsic
lifetime and homogeneous and
inhomogeneous broadening can be
separated.

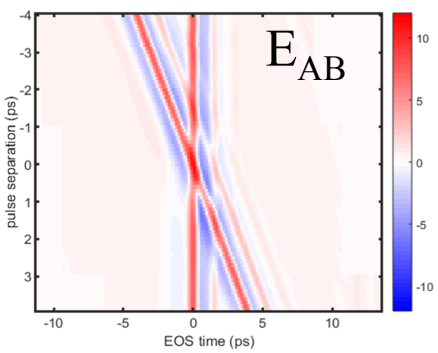
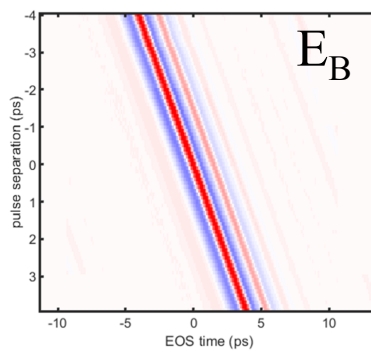
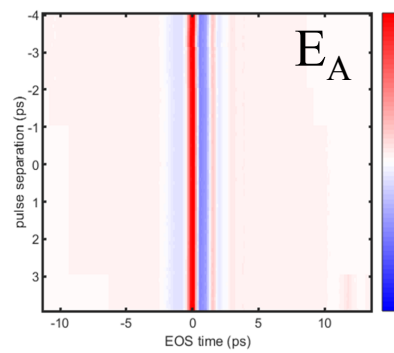
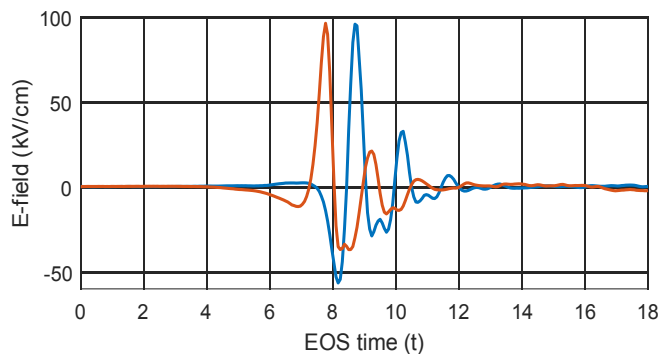
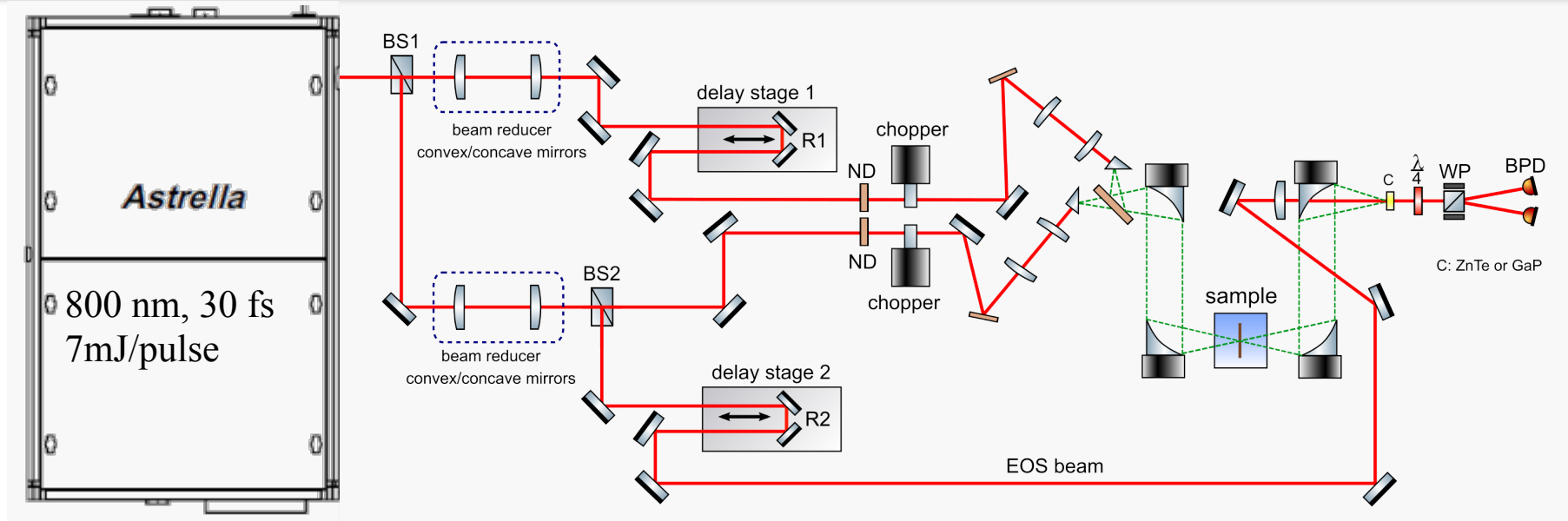


2D NMR Spectroscopy

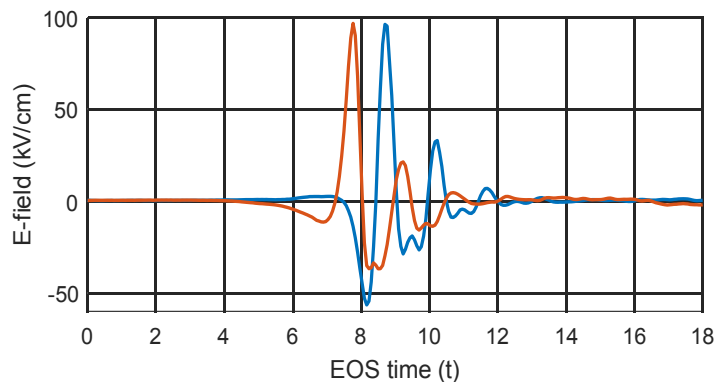
Nuclear Overhauser Effect Spectroscopy
(NOESY) spectra of codeine



2D THz Coherent Spectroscopy

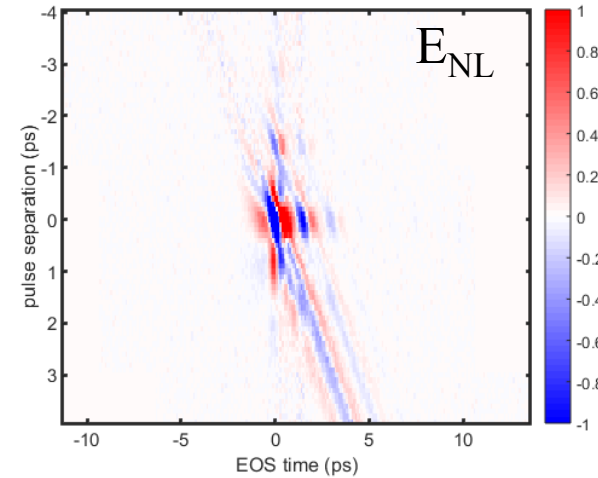
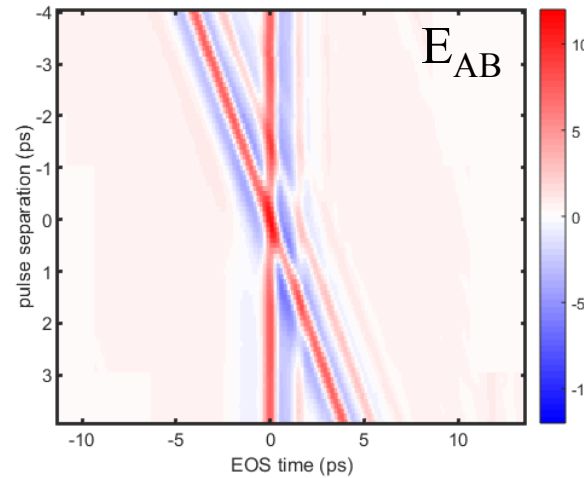
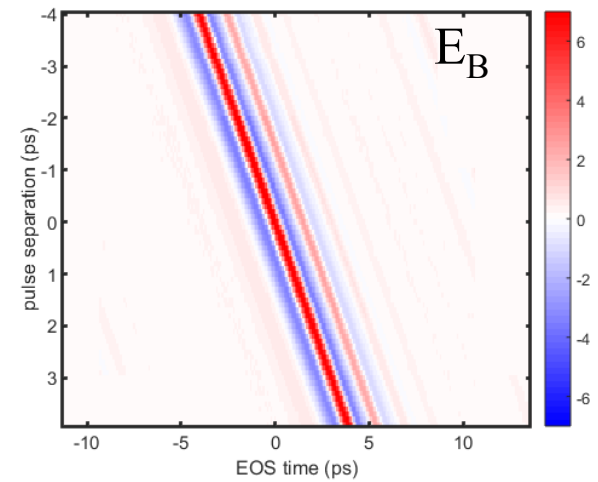
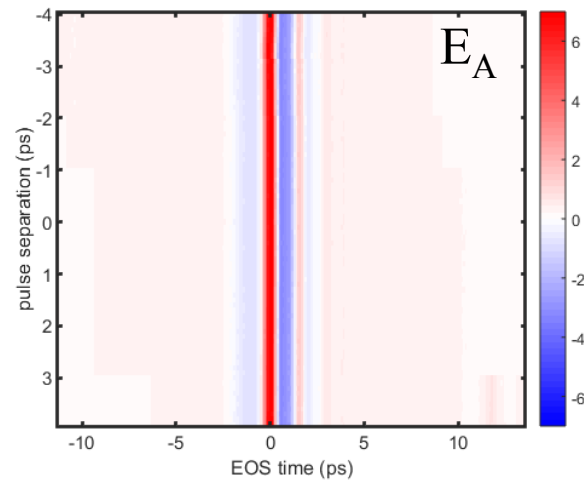


2D THz Coherent Spectroscopy



Excite with two pulses. Slide through each other in time. Look at difference signal with both together from each separately.

$$E_{NL} = E_{AB} - E_A - E_B$$



Third order non-linear response

$$E_j = E \cos(\omega t) = (E e^{i\omega_j t} + E^* e^{-i\omega_j t})/2$$

$$\chi_{ijkl} = \chi(\omega_i; \omega_j, \omega_k, \omega_l)$$

$$\Delta P_i = \chi_{ijjk}[(E_j)^2 E_k] + \chi_{ijkk}[E_j (E_k)^2]$$

$$\begin{aligned} \propto \chi_{ijjk} & [(EE)E e^{i(2\omega_j + \omega_k)t} + (EE^*)E e^{i(\omega_j - \omega_j + \omega_k)t} + (E^*E)E e^{i(-\omega_j + \omega_j + \omega_k)t} + (E^*E^*)E e^{i(-2\omega_j + \omega_k)t} + \\ & (EE)E^* e^{i(2\omega_j - \omega_k)t} + (EE^*)E^* e^{i(\omega_j - \omega_j - \omega_k)t} + (E^*E)E^* e^{i(-\omega_j + \omega_j - \omega_k)t} + (E^*E^*)E^* e^{i(-2\omega_j - \omega_k)t}] + \\ \chi_{ijkk} & [E(EE)e^{i(\omega_j + 2\omega_k)t} + E(EE^*)e^{i(\omega_j + \omega_k - \omega_k)t} + E(E^*E)e^{i(\omega_j - \omega_k + \omega_k)t} + E(E^*E^*)e^{i(\omega_j - 2\omega_k)t} + \\ & E^*(EE)e^{i(-\omega_j + 2\omega_k)t} + E^*(EE^*)e^{i(-\omega_j + \omega_k - \omega_k)t} + E^*(E^*E)e^{i(-\omega_j - \omega_k + \omega_k)t} + E^*(E^*E^*)e^{i(-\omega_j - 2\omega_k)t}] \end{aligned}$$

3HG

PP

PP

2Q

$$\begin{aligned} \Delta P_i \propto \chi_{ijjk} & [(EE)E e^{i(2\omega_j + \omega_k)t} + (EE^*)E e^{i(\omega_j - \omega_j + \omega_k)t} + (E^*E)E e^{i(-\omega_j + \omega_j + \omega_k)t} + (EE)E^* e^{i(2\omega_j - \omega_k)t}] + \\ & \chi_{ijkk} [E(EE)e^{i(\omega_j + 2\omega_k)t} + E(EE^*)e^{i(\omega_j + \omega_k - \omega_k)t} + E(E^*E)e^{i(\omega_j - \omega_k + \omega_k)t} + E(E^*E^*)e^{i(-\omega_j + 2\omega_k)t}] + c.c. \end{aligned}$$

3HG

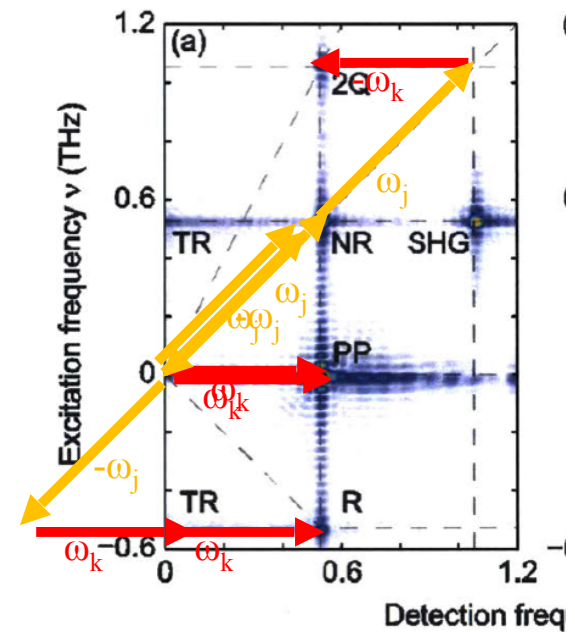
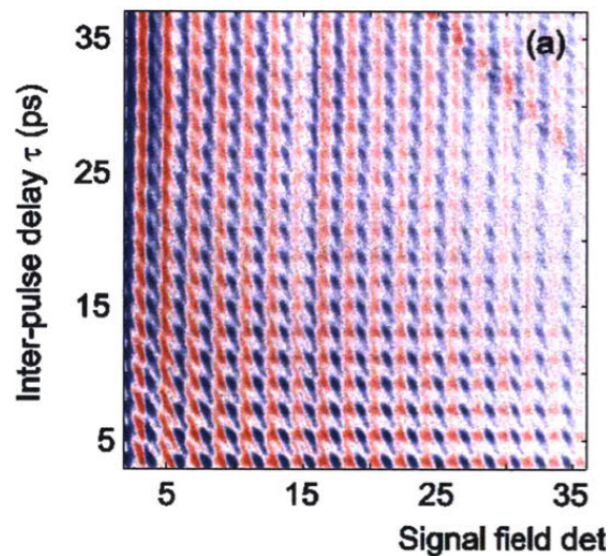
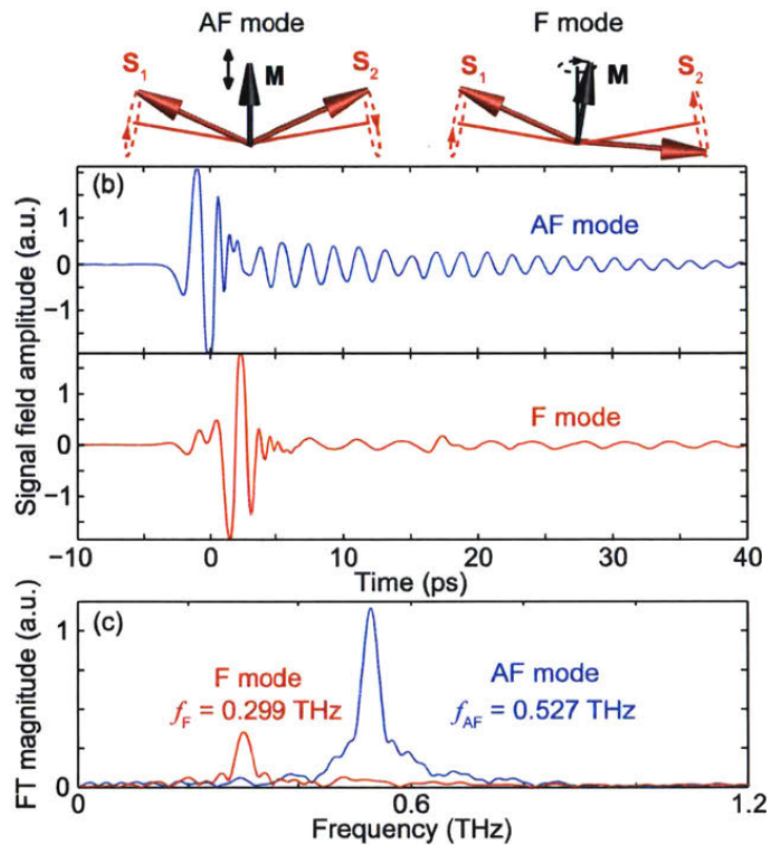
NR

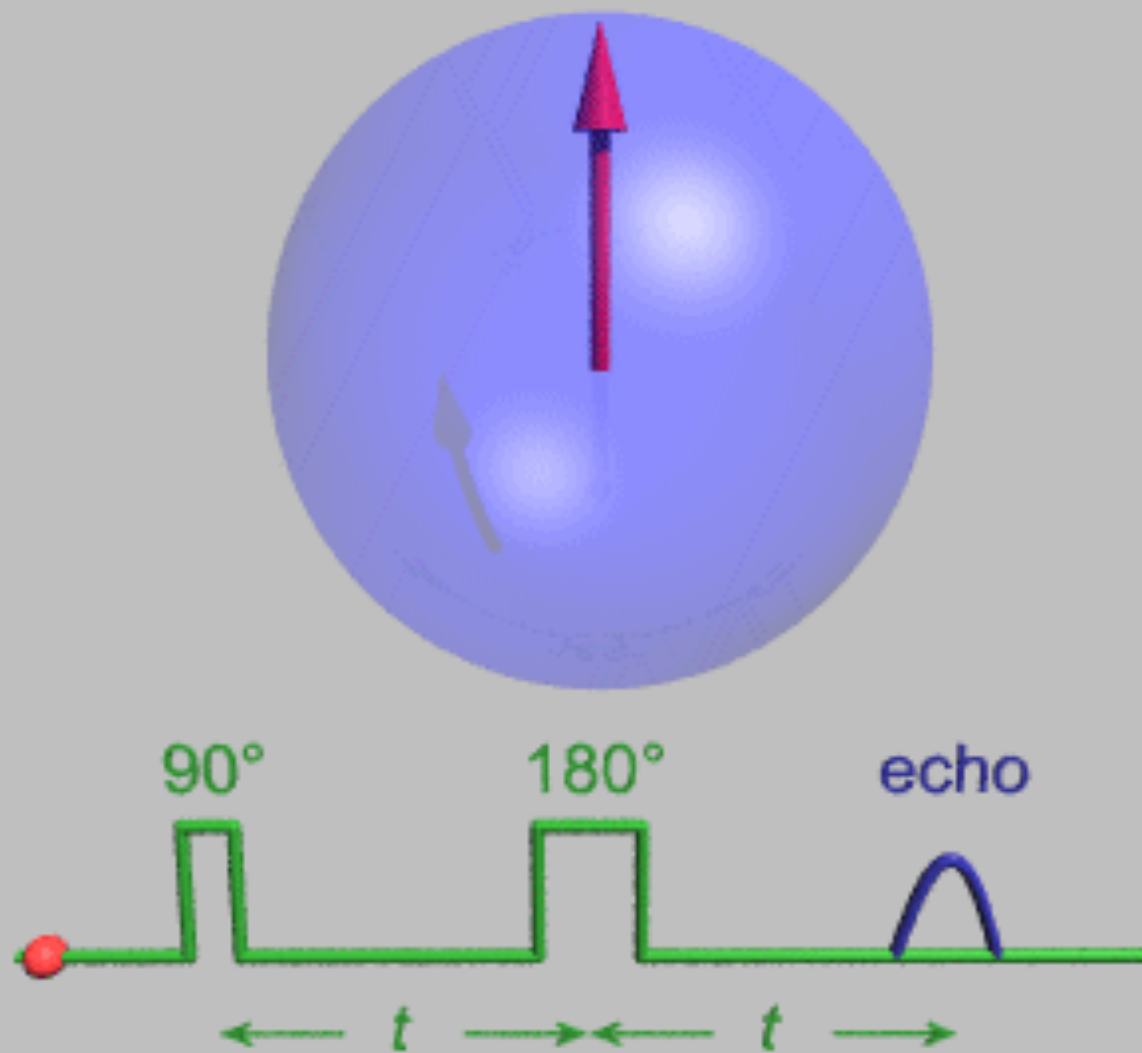
NR

R

Example: Magnons in AFM

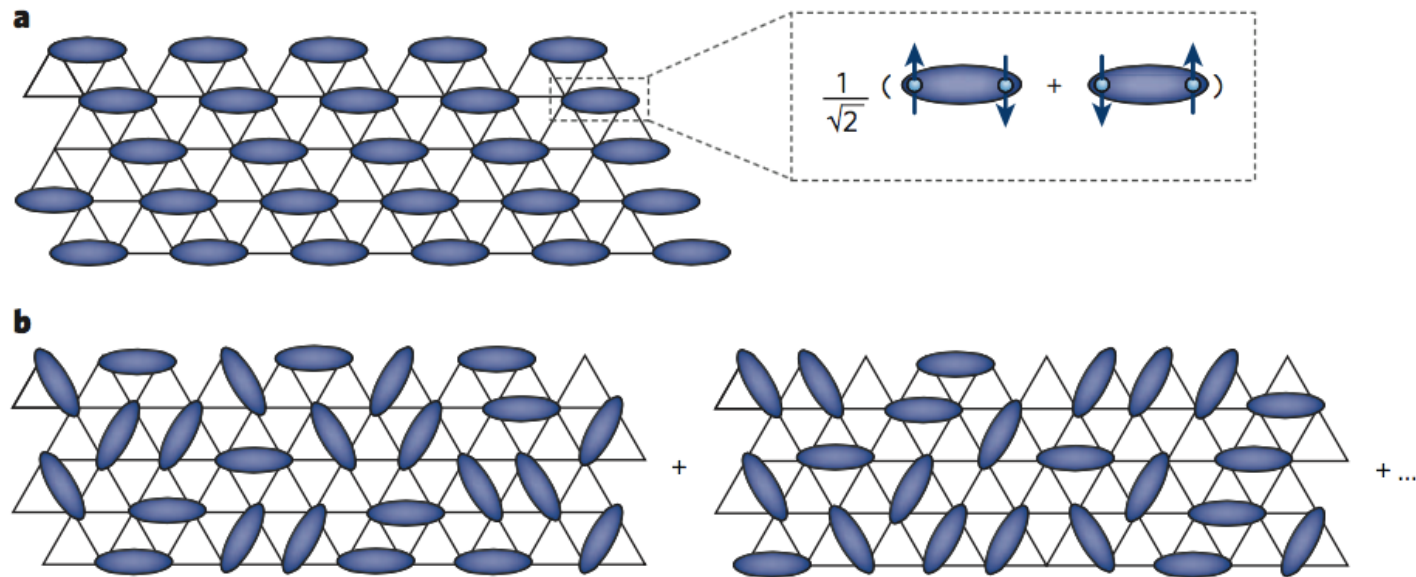
- YFeO₃ : Canted Anti-ferromagnet

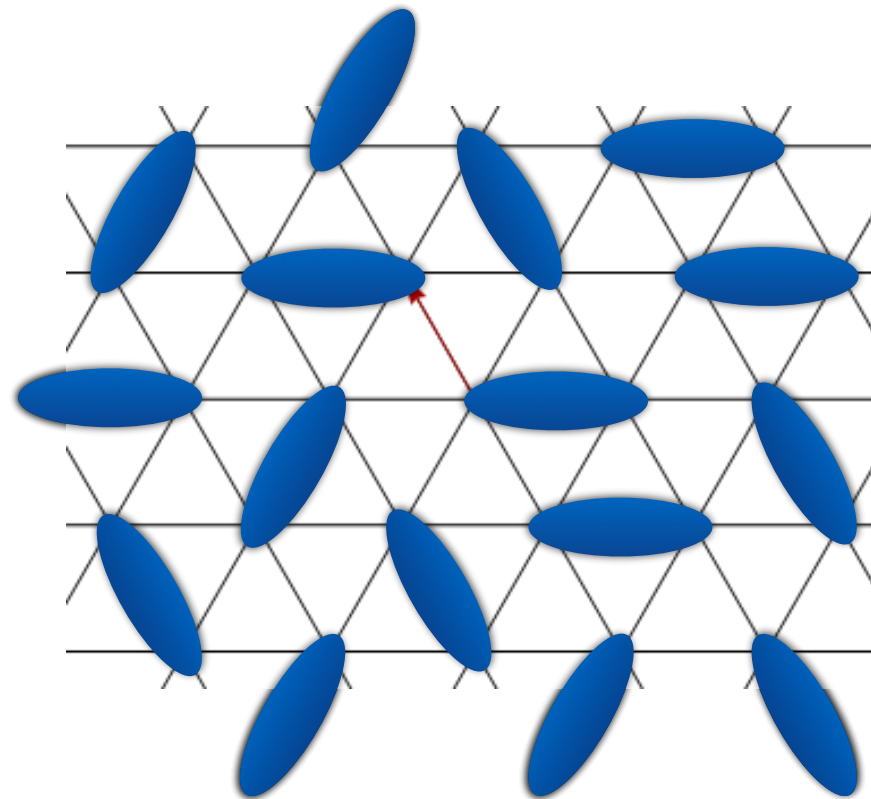


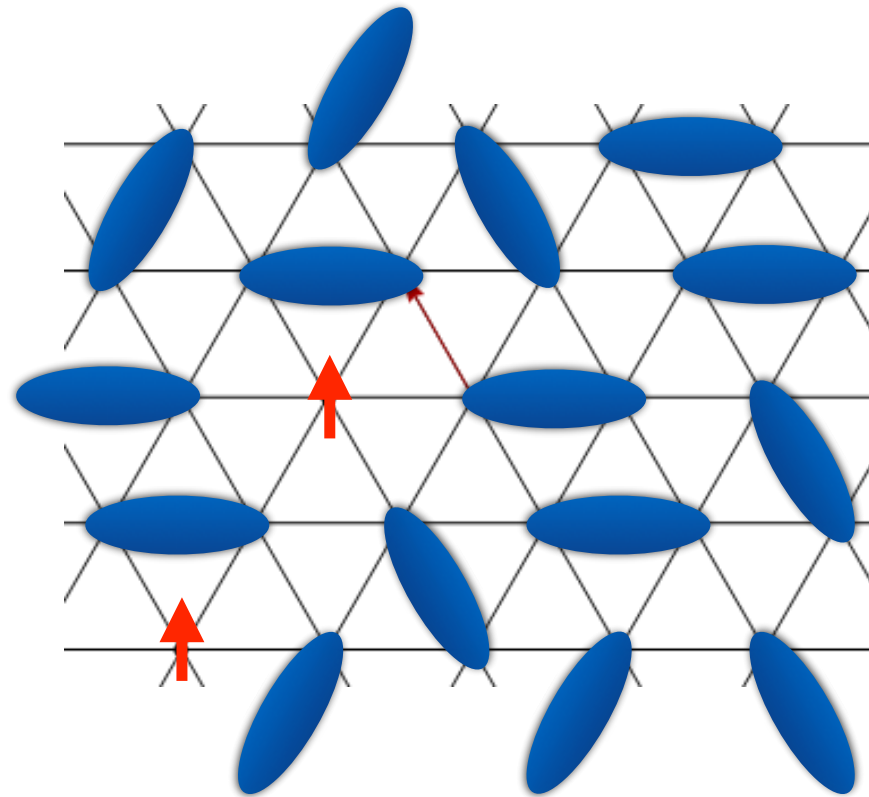


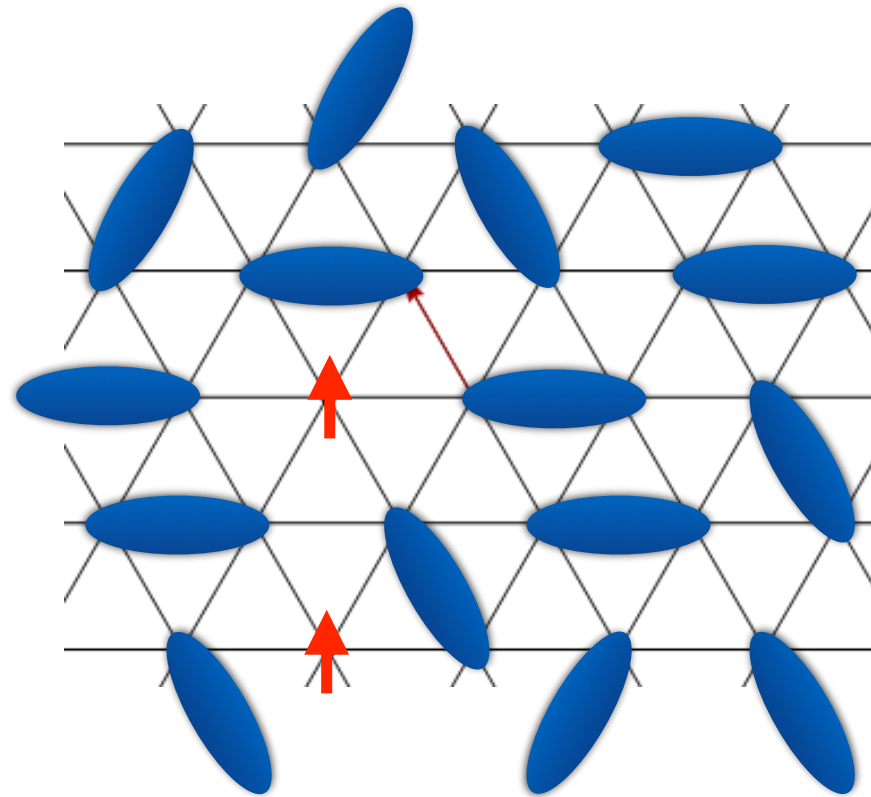
Spin systems with fractionalization

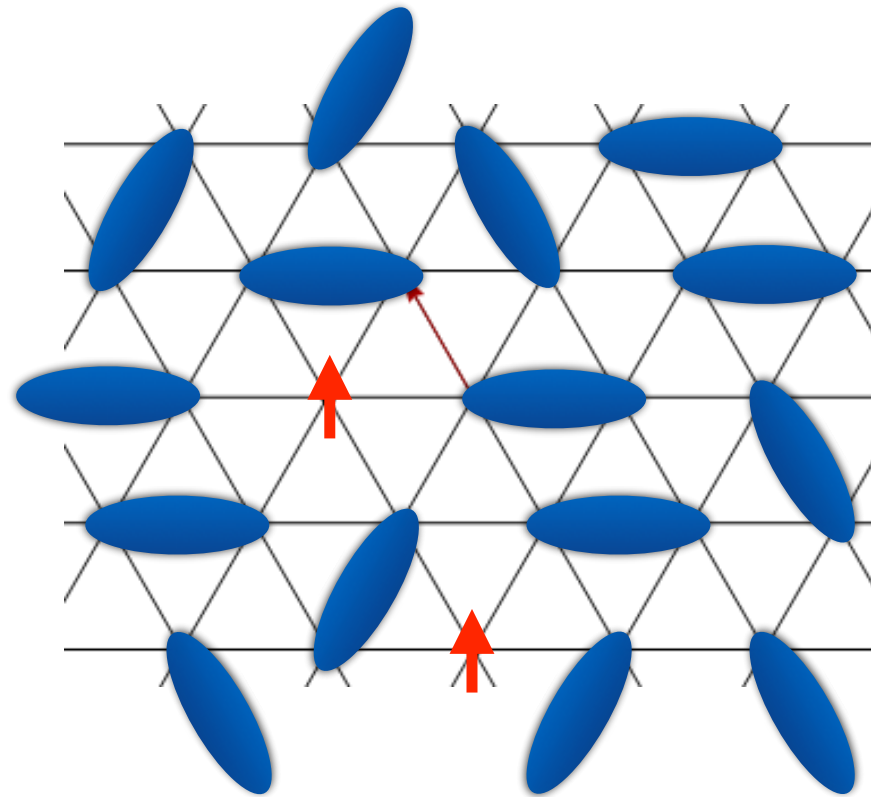
Spin singlet formation in a spin liquid

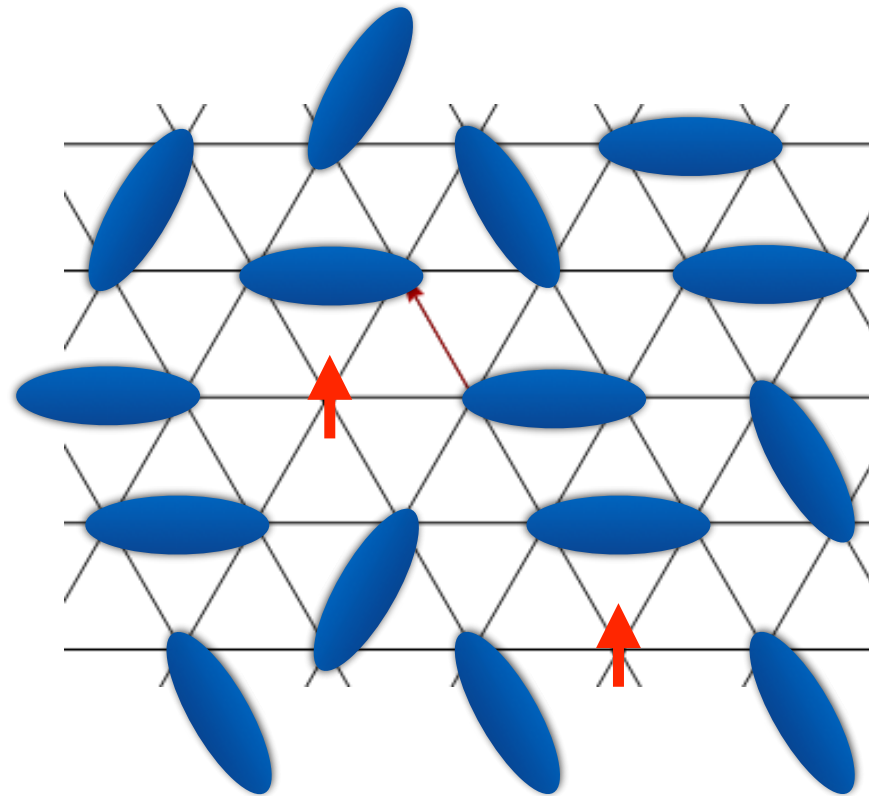


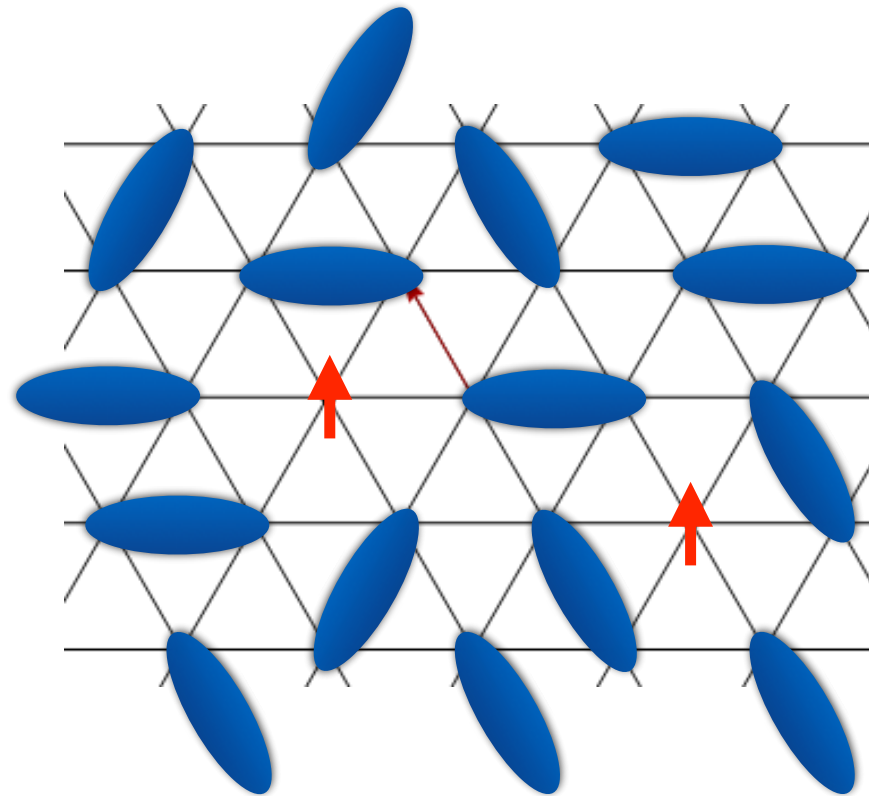


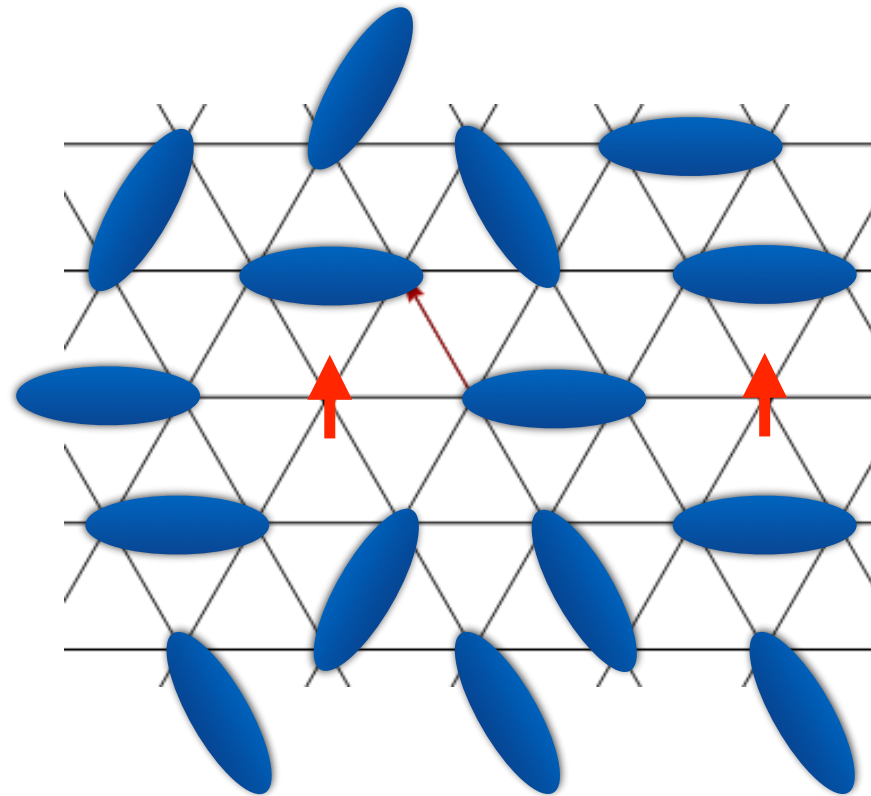


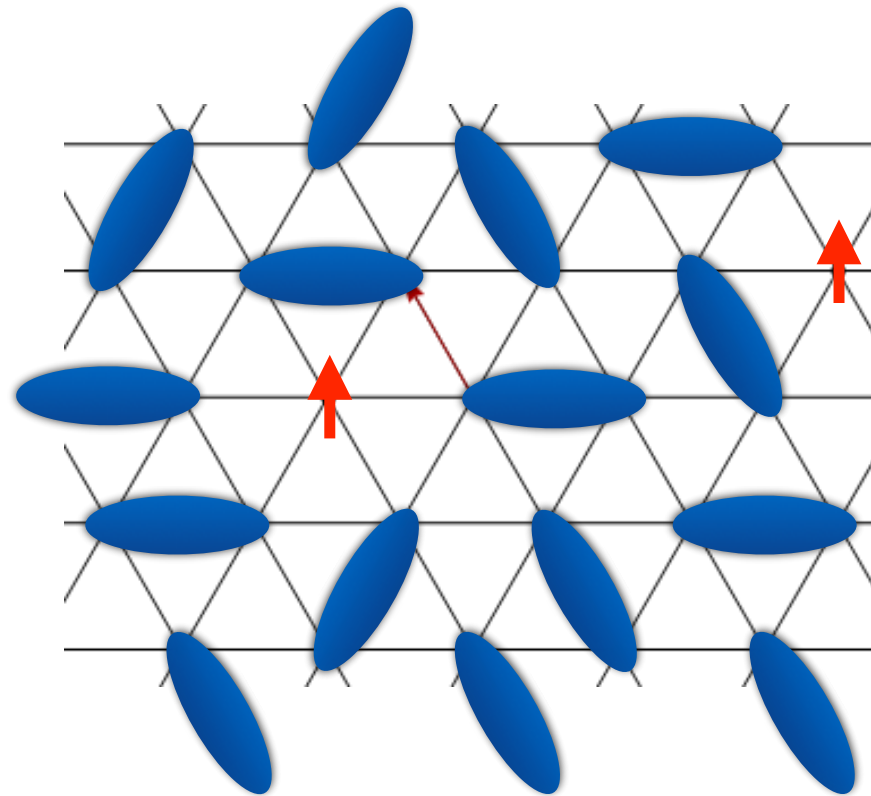






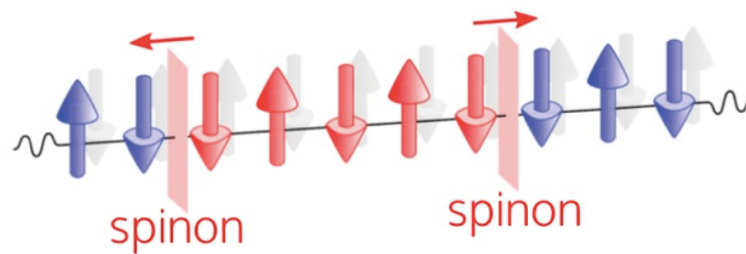
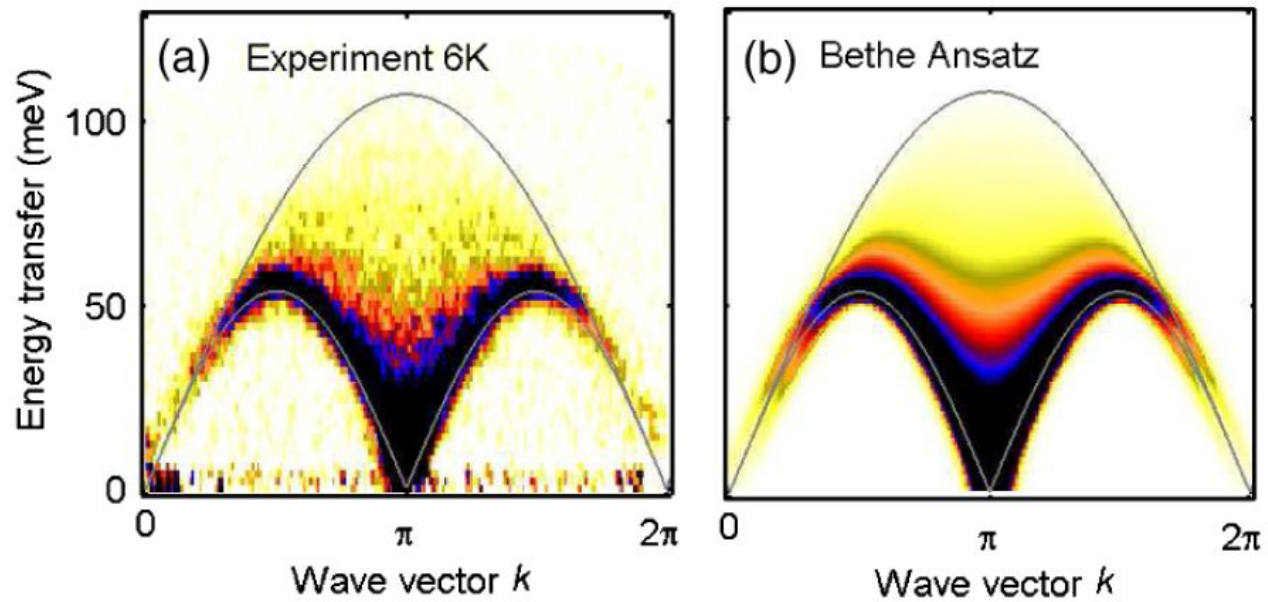






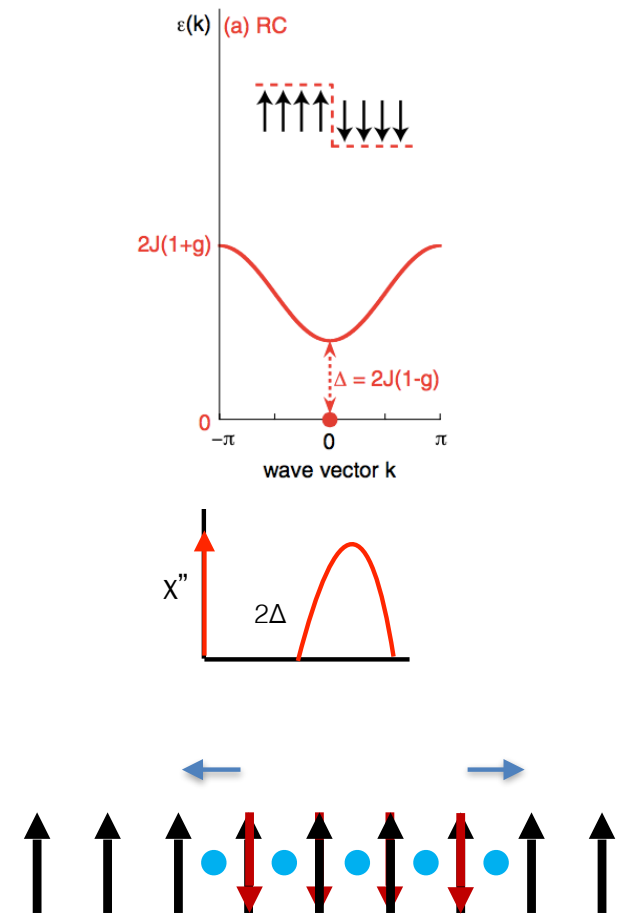
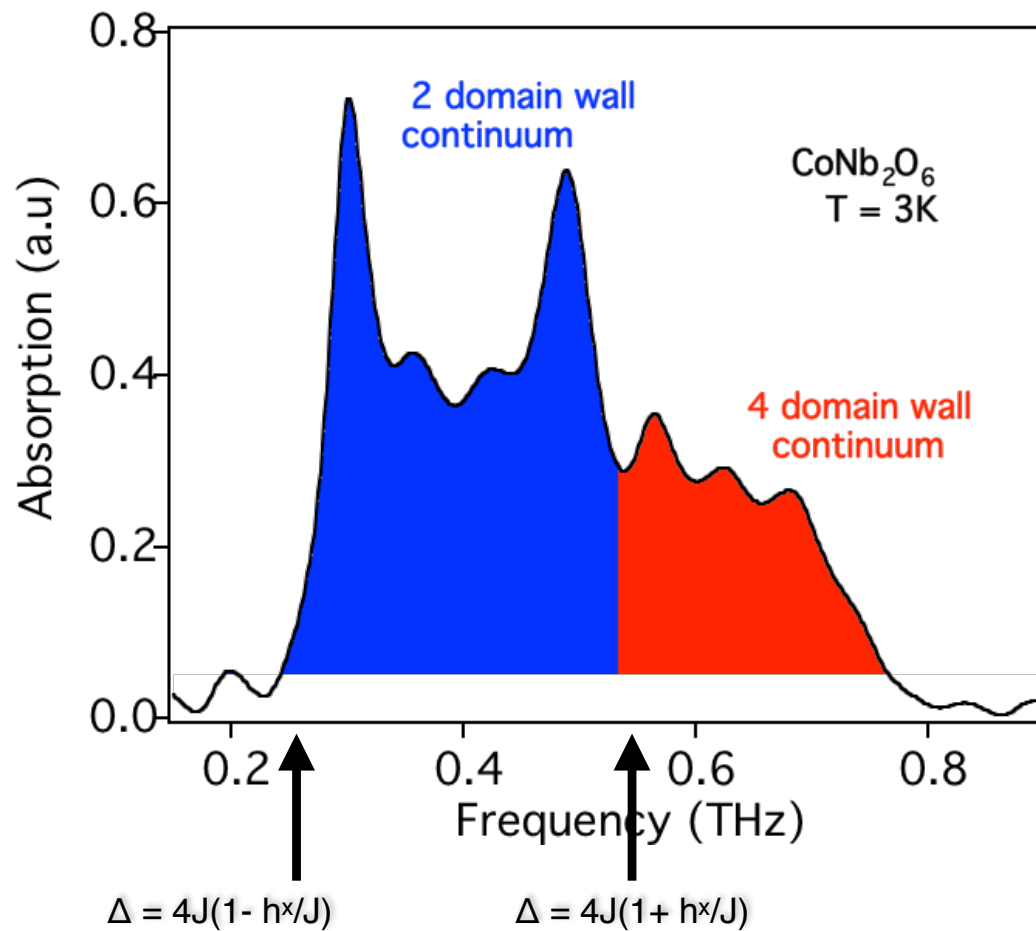
Spinon Continuum in 1D KCuF_3

B. Lake et al. (2013)

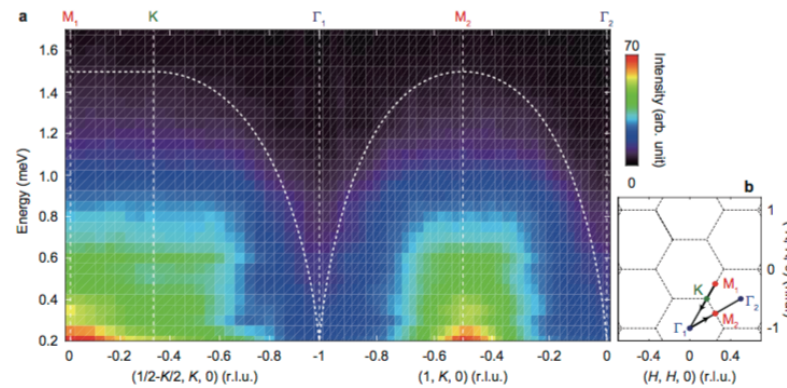


Mourigal et al. (2013)

Fractionalization: 1D Ising chain in small transverse field



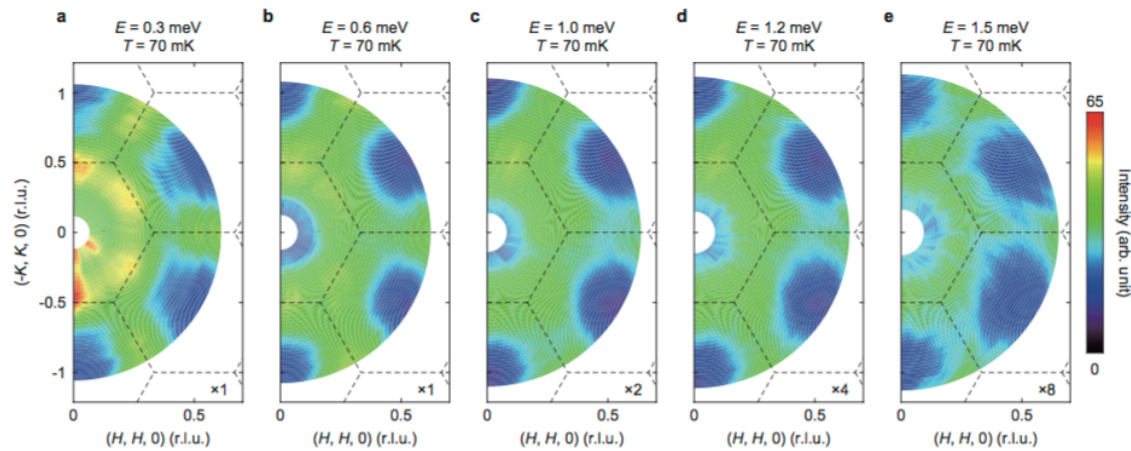
Purported Spinon Continuum in higher dimension



YbMgGaO₄

Evidence for spinon continuum less convincing in higher D.

Broad spectral shapes hard to distinguish from disorder



Shen et al. 2017

Resolving Continua of Fractional Excitations by Spinon Echo in THz 2D Coherent Spectroscopy

Yuan Wan^{1,*} and N. P. Armitage^{2,3,†}

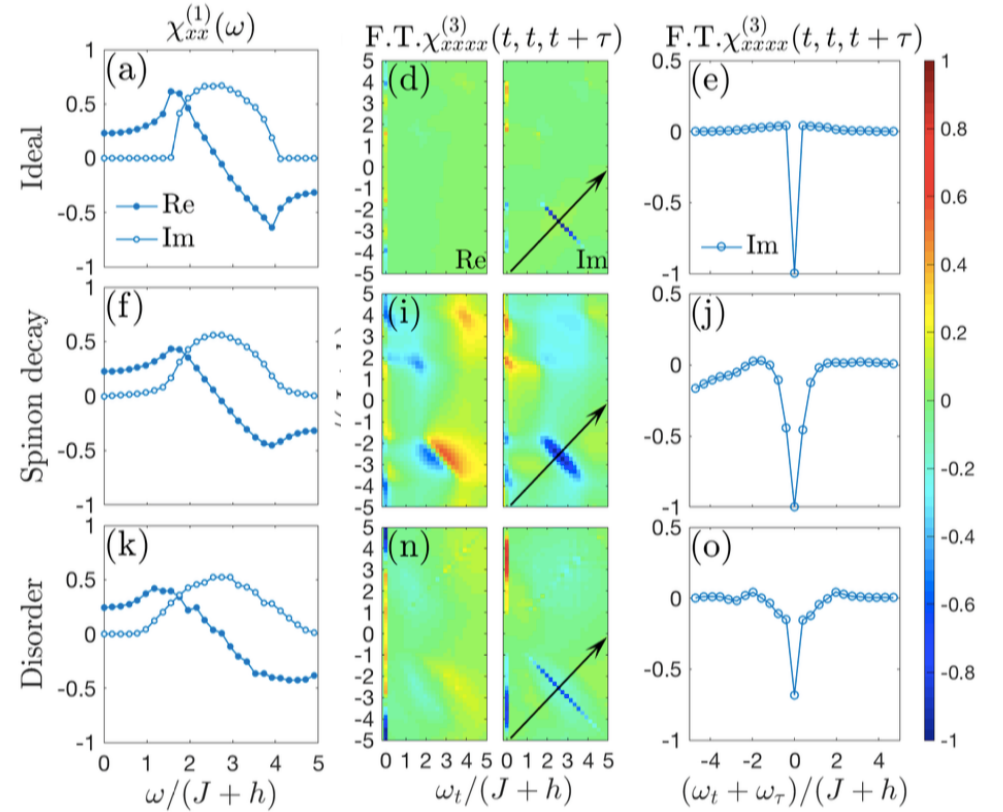
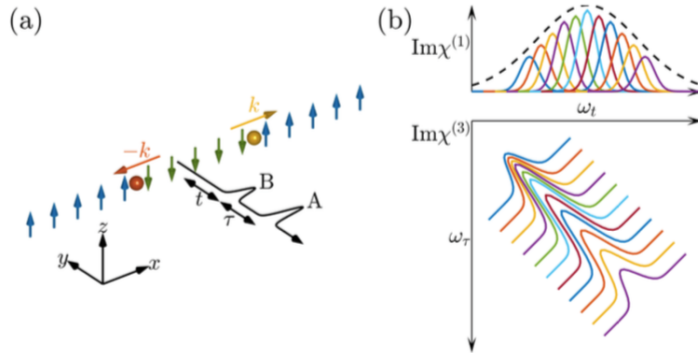
¹*Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

²*Institute for Quantum Matter and Department of Physics and Astronomy, Johns Hopkins University, Baltimore, Maryland 21218, USA*

³*Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581, Japan*

$$H = -J \left(\sum_{n=1}^{L-1} \sigma_n^z \sigma_{n+1}^z + \eta \sigma_L^z \sigma_1^z \right) - h \sum_n \sigma_n^x$$

Simplest model for fractionalization.
 Ising chain in transverse field. Can be
 mapped to model of non-interacting
 particles.



Electron Glasses

Lightly doped Si

Si:P @ 39% of n_c

$$n_P \sim 1.8 \times 10^{18}/\text{cm}^3$$

$$\xi \sim 13 \text{ nm}$$

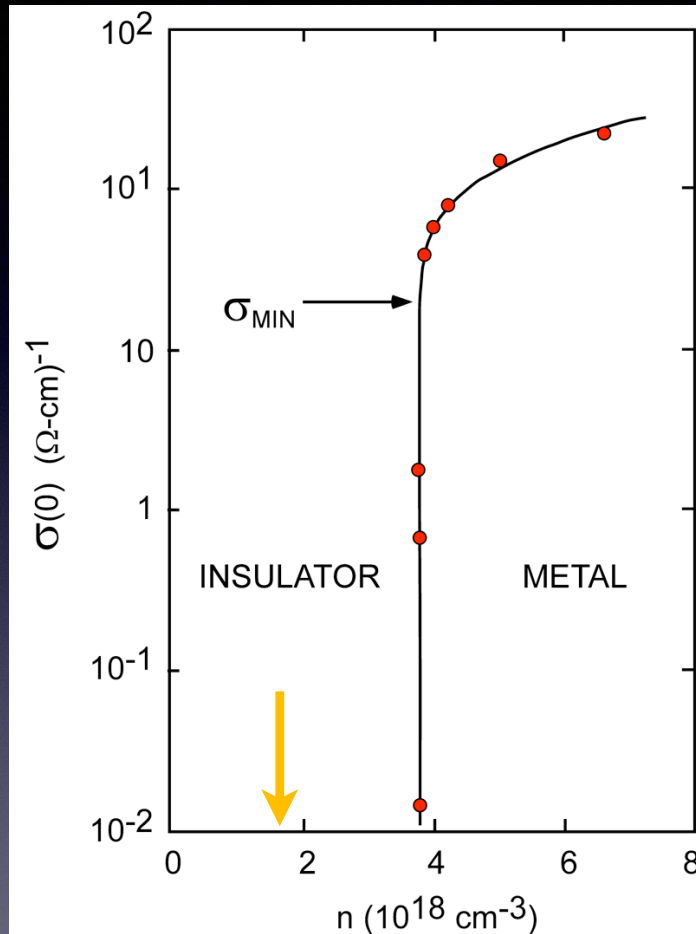
$$n_P^{-1/3} \sim 9 \text{ nm}$$

$$r_{\text{Bohr}} \sim 2.5 \text{ nm}$$

$$\epsilon \sim 14 \quad (\epsilon_{\text{Si}} \sim 11.7)$$

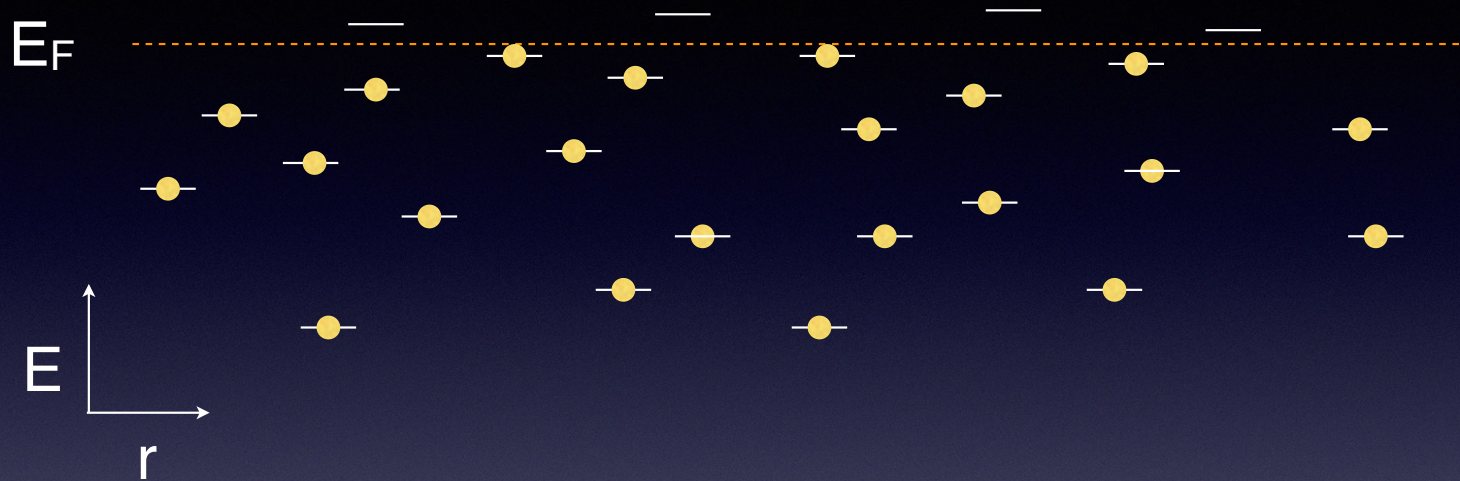
$$\xi > n_P^{-1/3}$$

Samples previously used for studies of
DC hopping conductivity and AC
conductivity in phononless regime



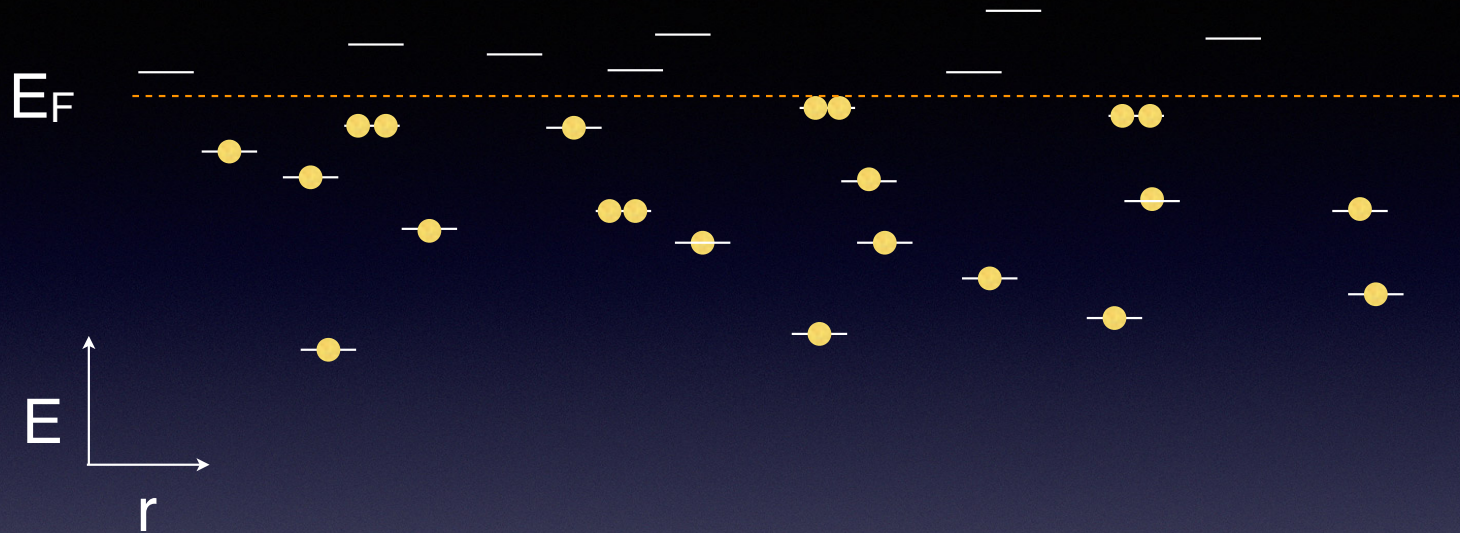
Rosenbaum et al. PRL 1983

Electron Glasses



Ensemble of orbitals localized in real space due to disorder — **Anderson insulator**

Electron Glasses

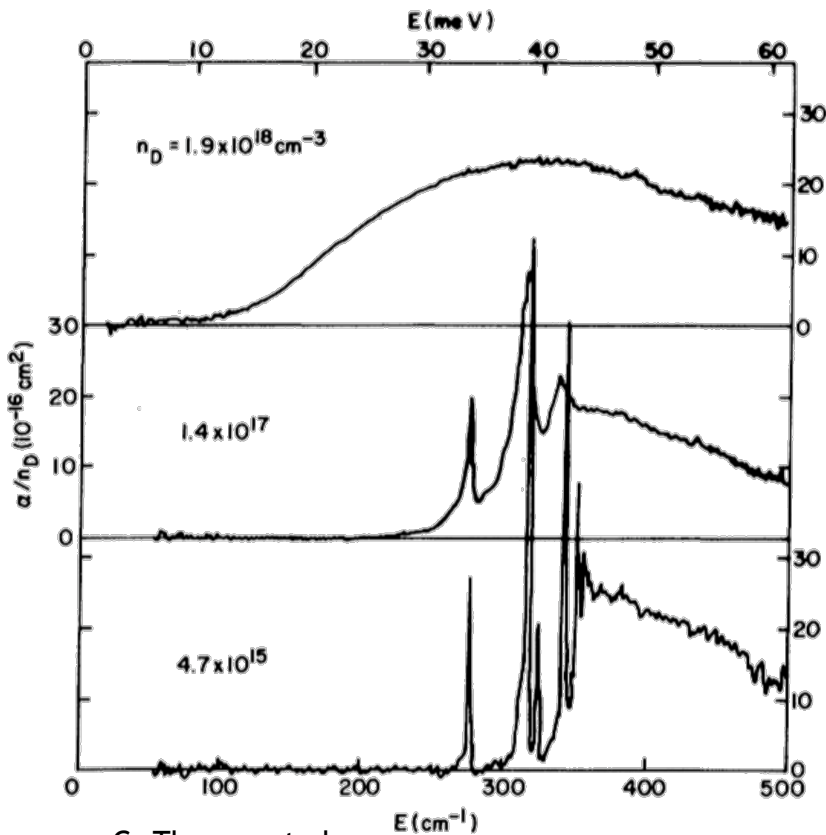


Insulators have long-range $1/r$ Coulomb interaction; no screening

Energy levels are not just a function of local disorder and site occupations; energy is a strong function of other occupations

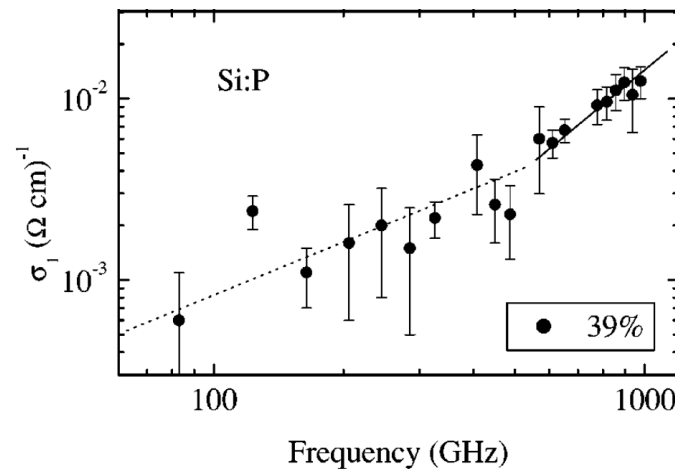
P doped Silicon

Phys. Rev. B 69, 014201 (2004)
Phys. Rev. B 23, 5472 (1981)



G. Thomas et al.

- At low ω optical conductivity is predicted (and found) to be power law (older work of NPA).
- “Resonant pair approximation” - understand as completely random ensemble of two level system.



Helgren, NPA, Gruner, 2002, 2004

$$\sigma = A\omega \left[\hbar\omega + \frac{e^2}{\epsilon_1 \langle r_\omega \rangle} \right]$$



Resonant pair approximation

Is the electron glass adiabatically connected to the Anderson insulator?

"(The) Fermi liquid theorem is a rigorous consequence of the exclusion principle, it happens because the phase space available for real interactions decreases so rapidly (as E^2 or T^2).

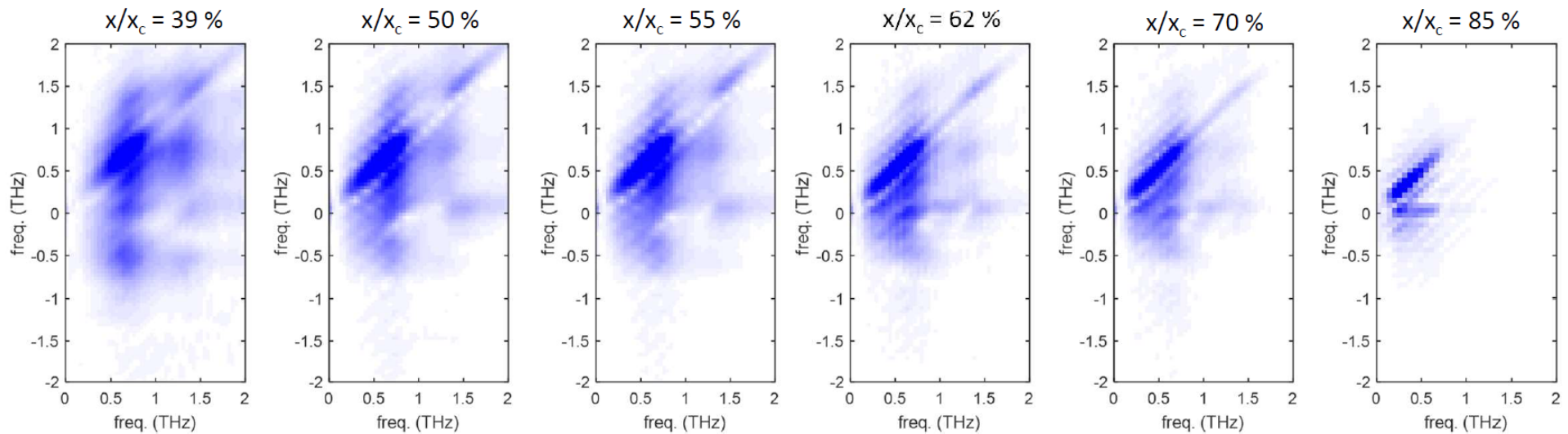
The theorem is equally true for the localized case: at sufficiently low temperatures or frequencies the non-interacting theory must be correct, even though the interactions are not particularly small or short range: thus the non-interacting theory is physically correct: the electrons can form a Fermi glass."

-P.W. Anderson, 1970

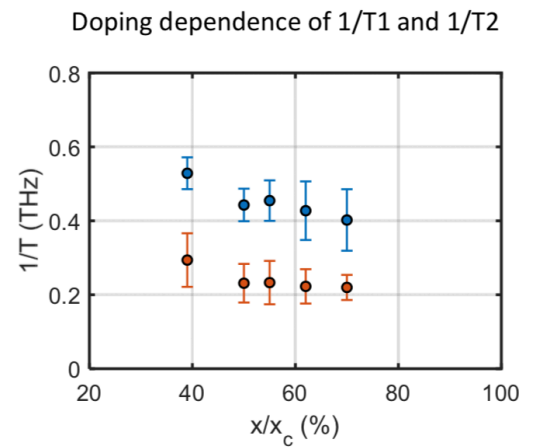
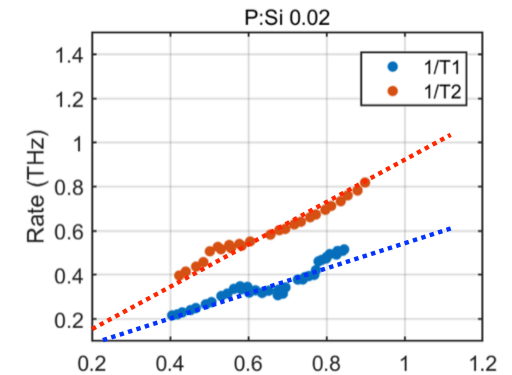
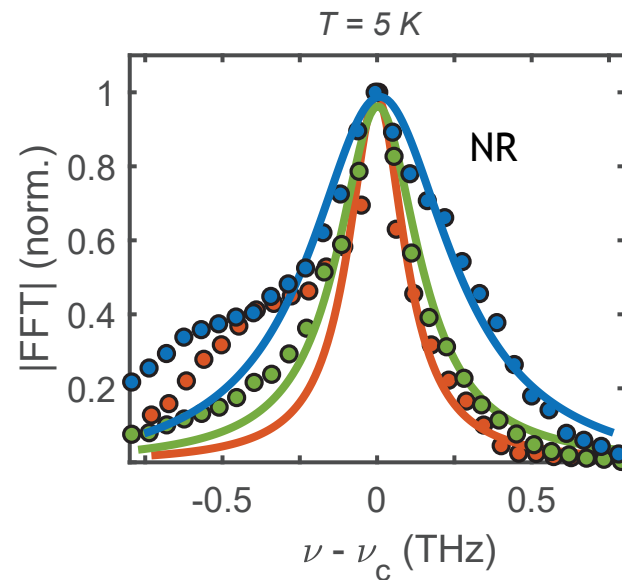
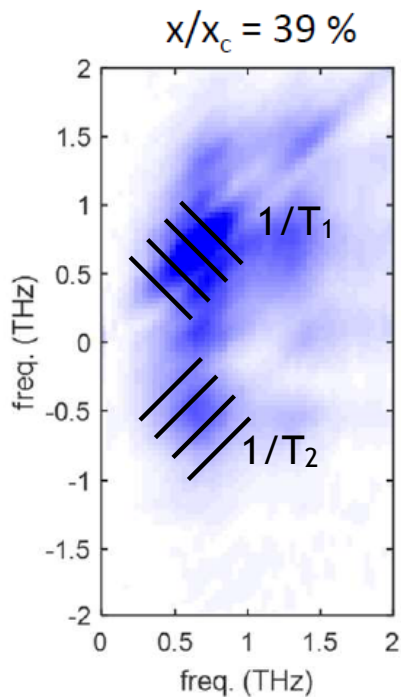
P doped Silicon: Doping dependence

Insulator

...Metal

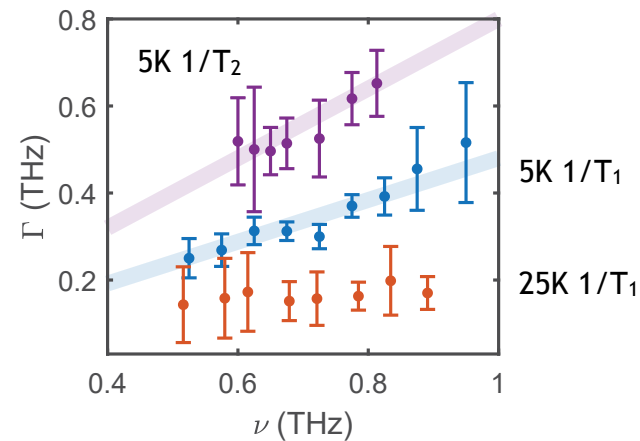
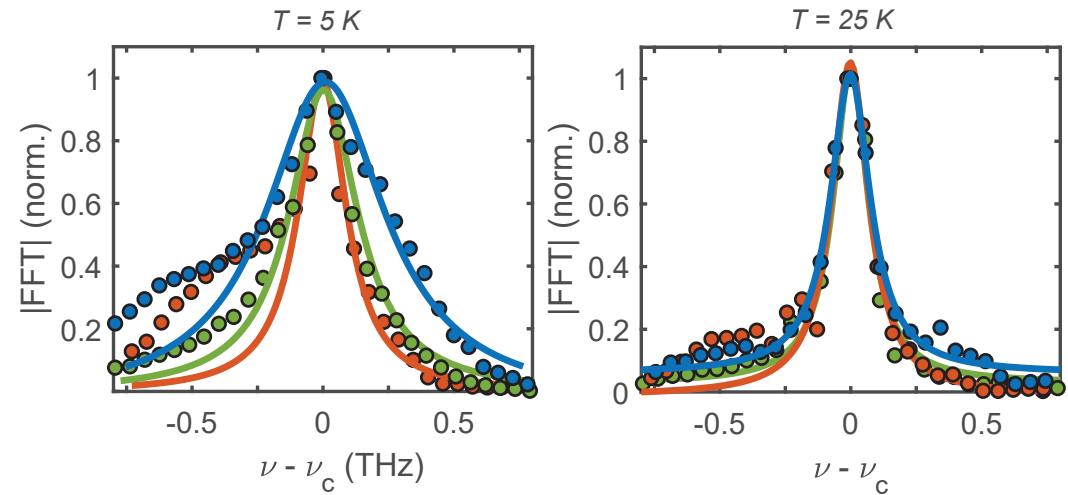
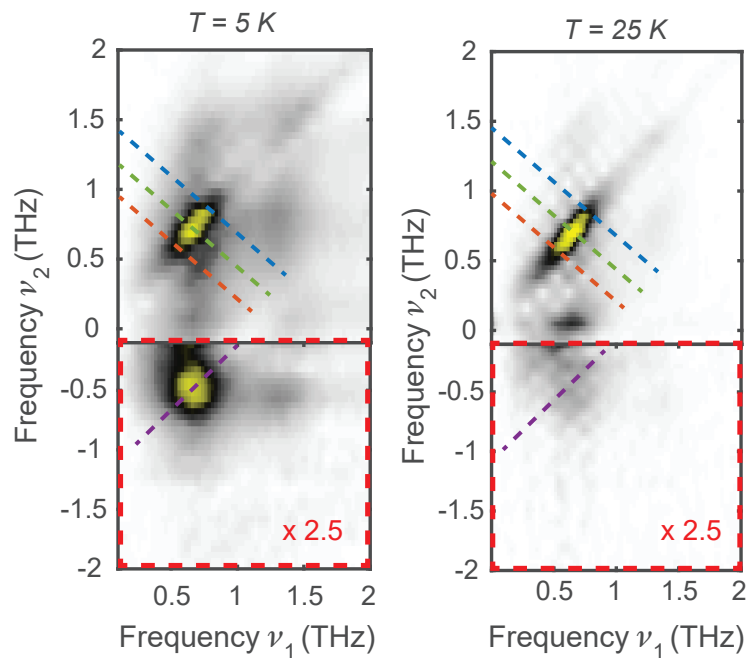


2D Terahertz spectroscopy on a localized electron glass system

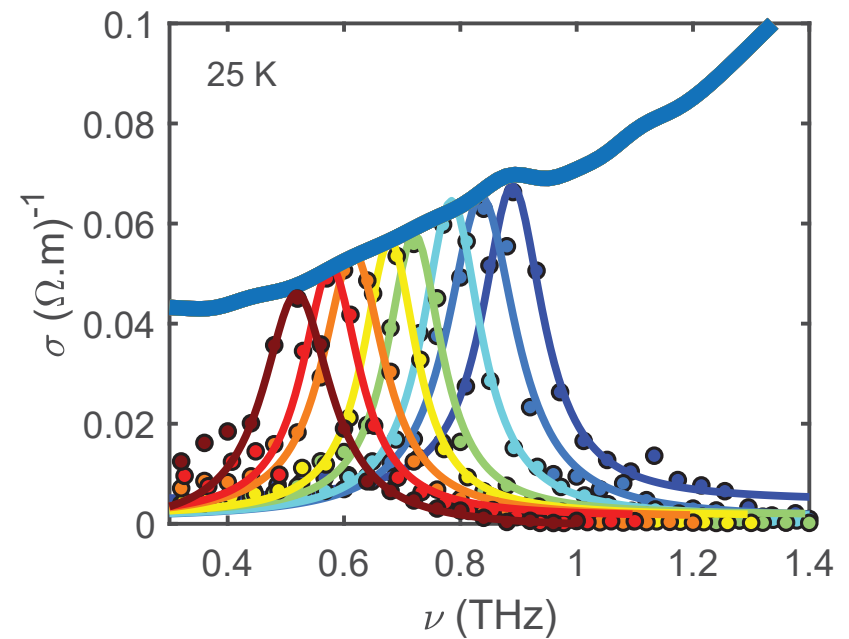
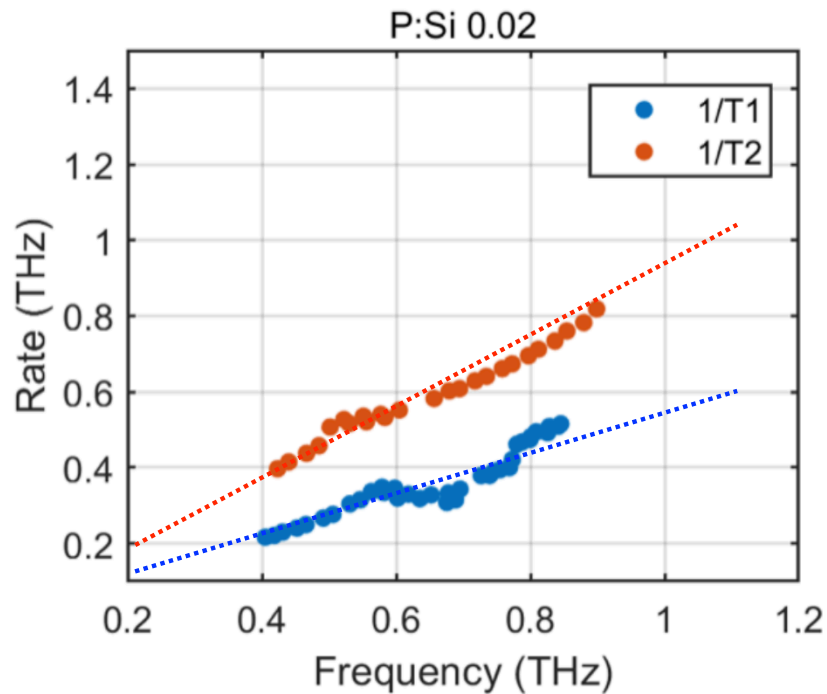


2D Terahertz spectroscopy on a localized electron glass system

Cuts along the NR quadrant

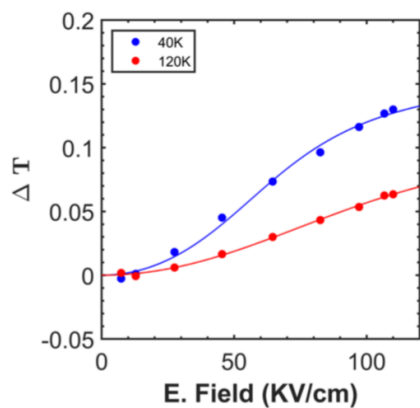
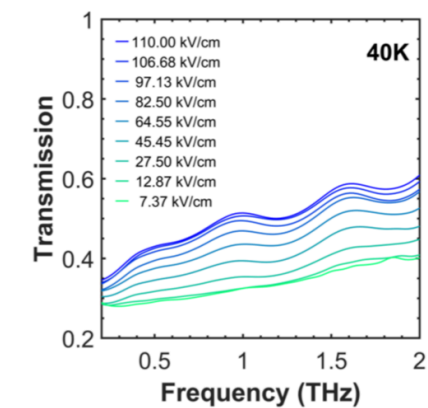


Sharp, but marginal excitations in an interacting localized system: “a non-Fermi glass”

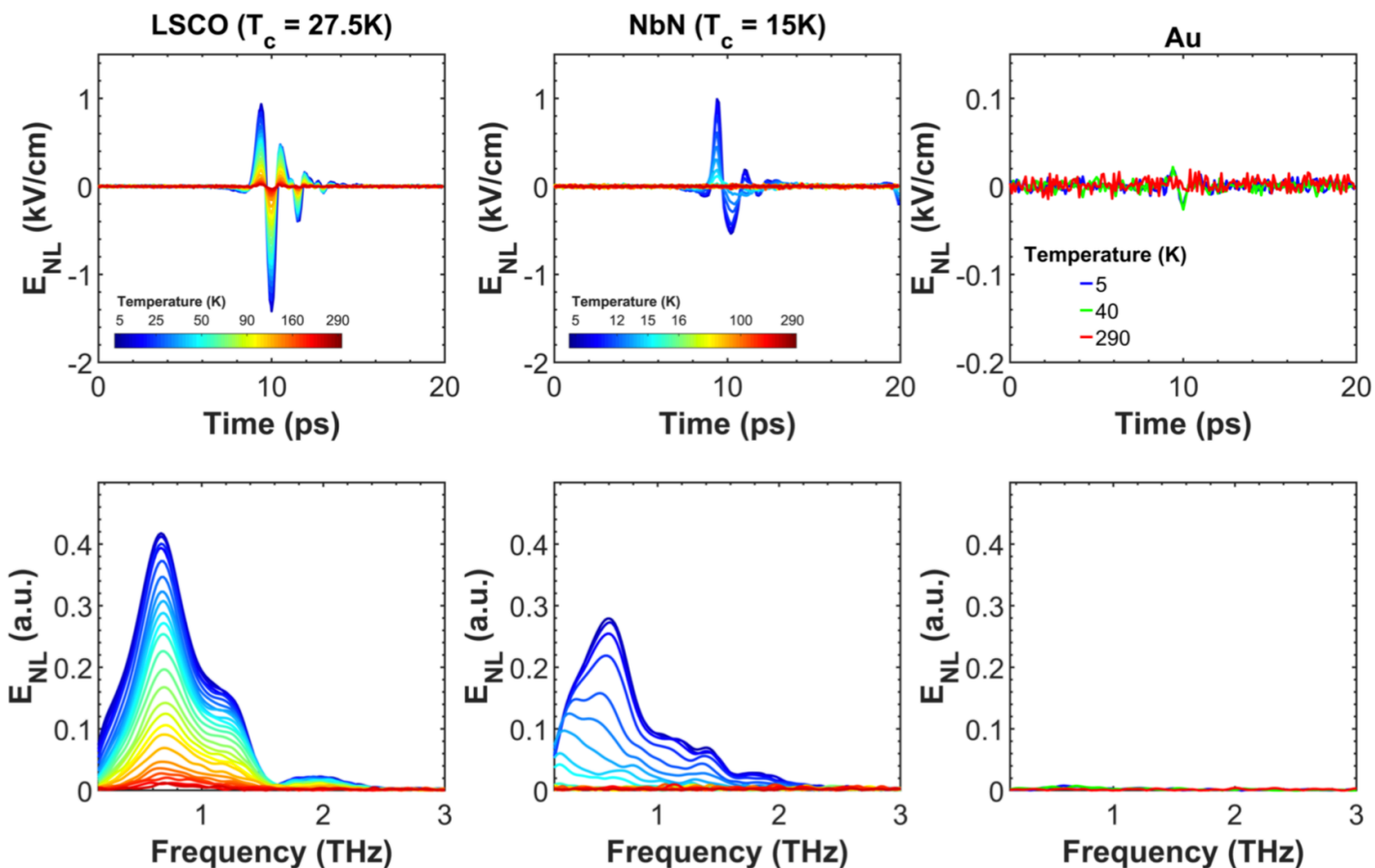




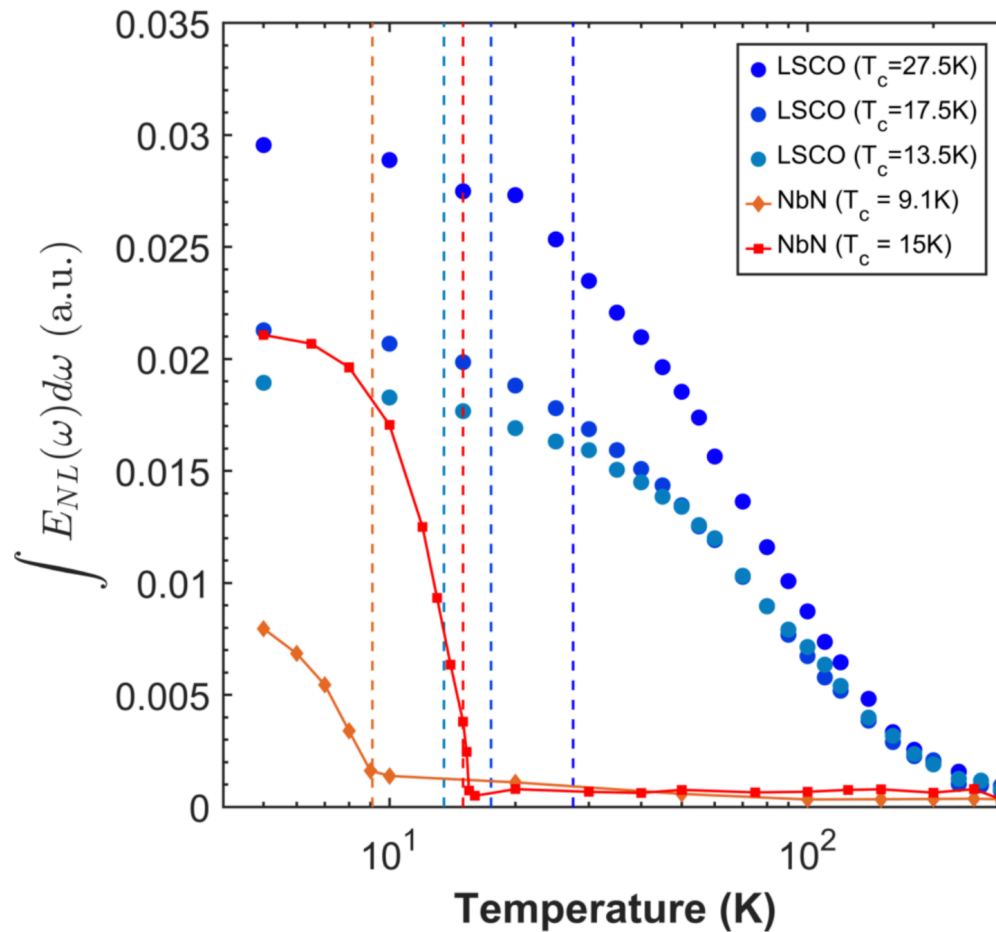
Optical response from one intense THz pulse



Non-Linear response from two overlapping intense THz pulses



Temperature dependence of THz nonlinearity spectroscopy contributions

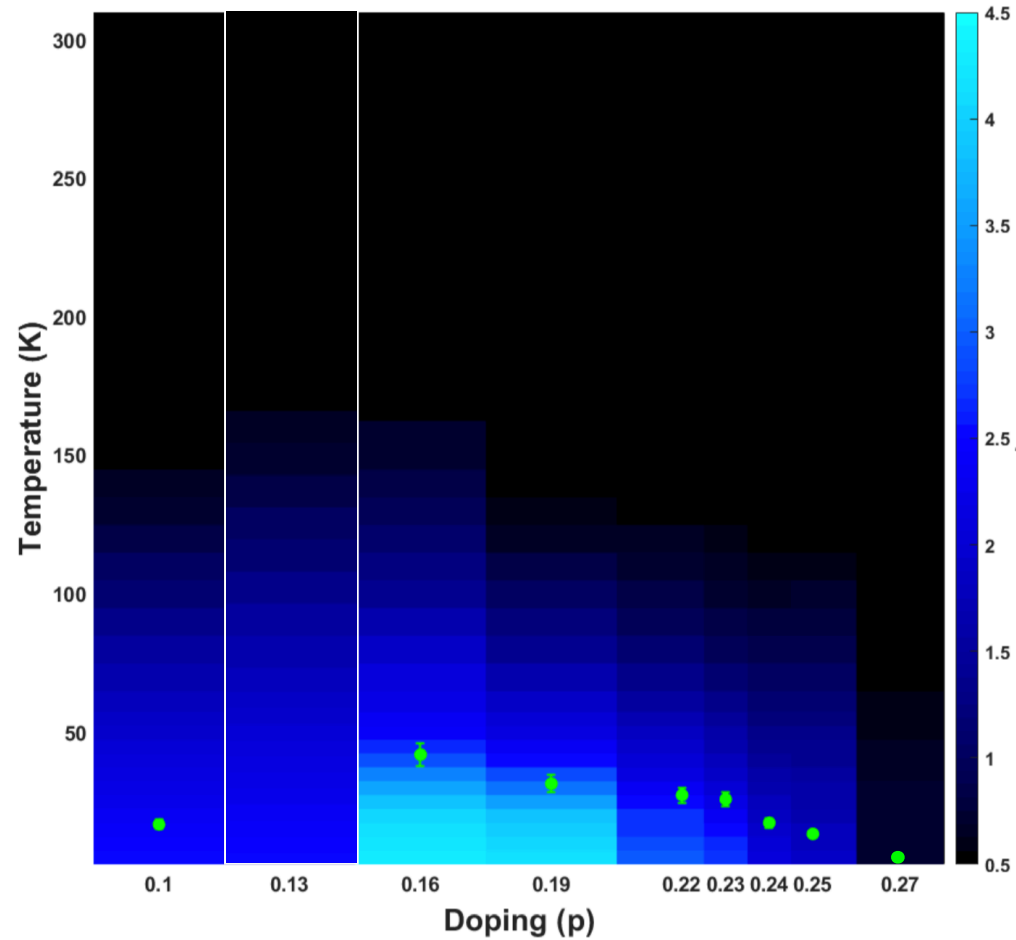


Large $\chi^{(3)}$ nonlinearity in superconductors is expected, but this is anomalous.

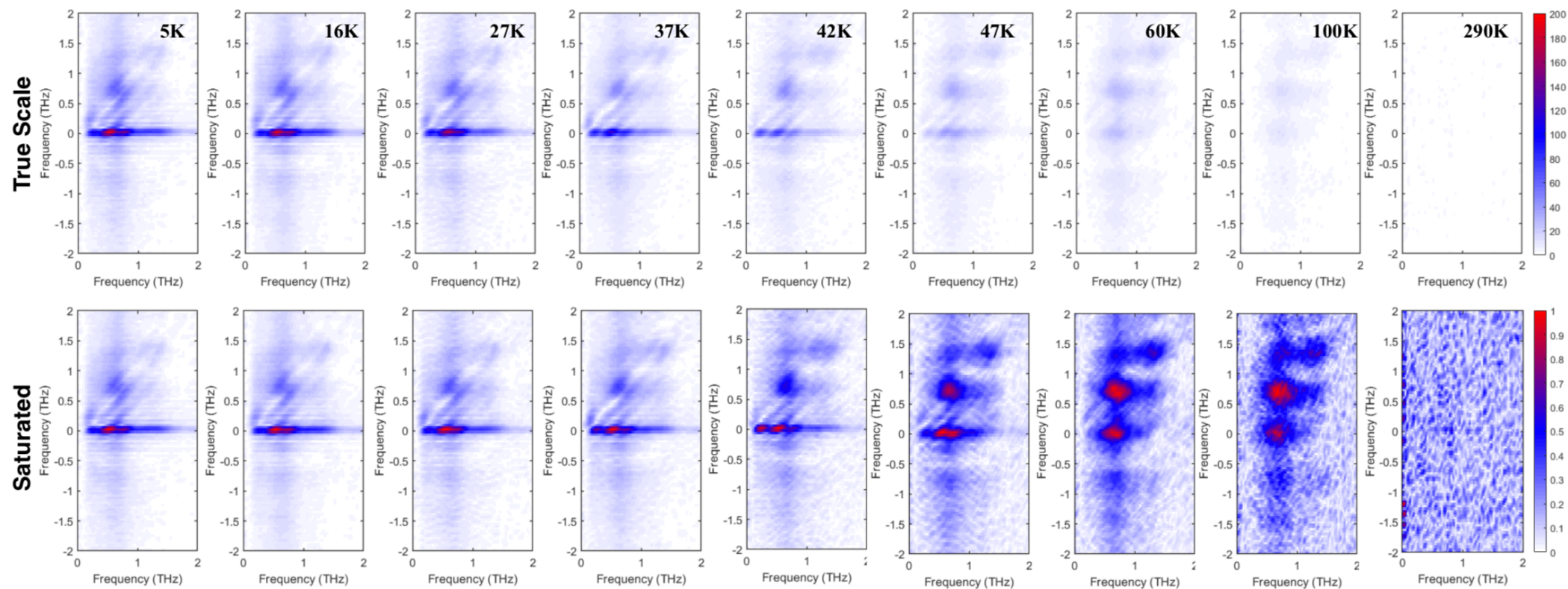
$$J_s = 2e\psi_\infty^2 \left(1 - \frac{m^* v_s^2}{2|\alpha|} \right) v_s$$

$$v_s \propto E$$

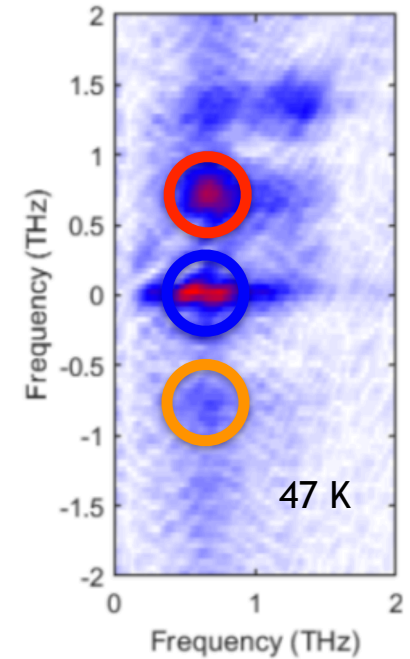
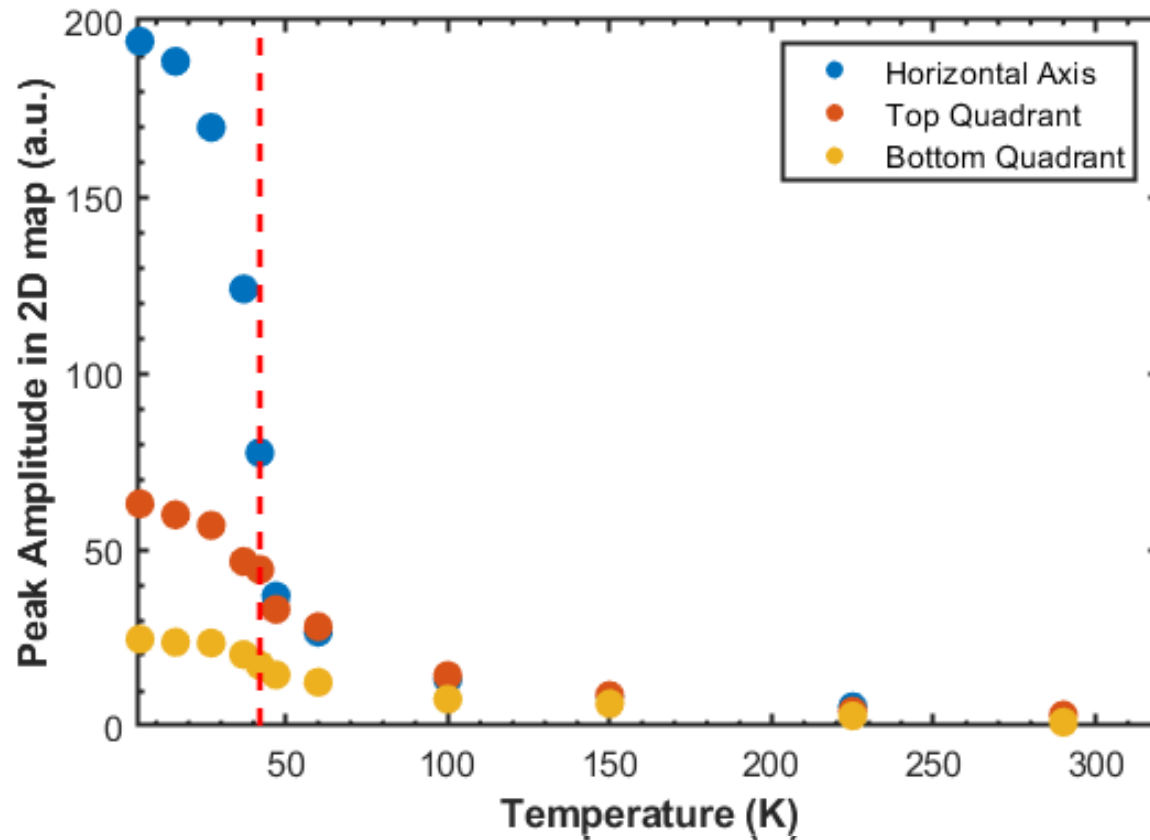
Temperature dependence of THz nonlinearity spectroscopy contributions



Temperature dependence of 2D spectroscopy contributions



Temperature dependence of 2D spectroscopy contributions



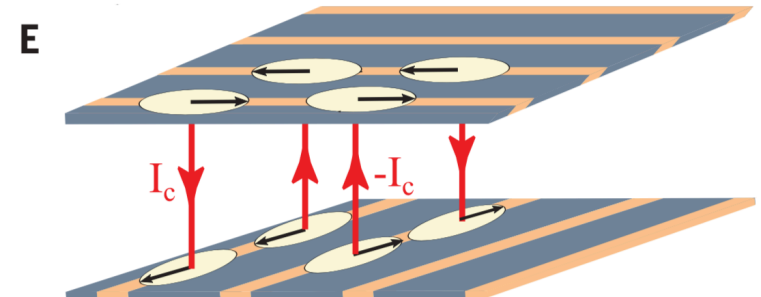
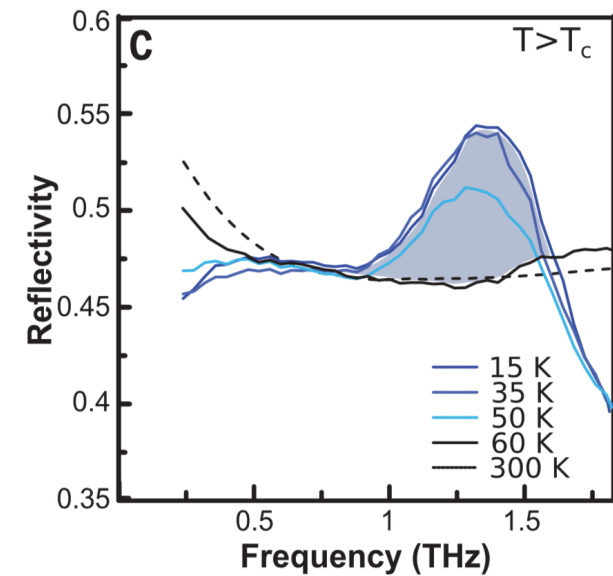
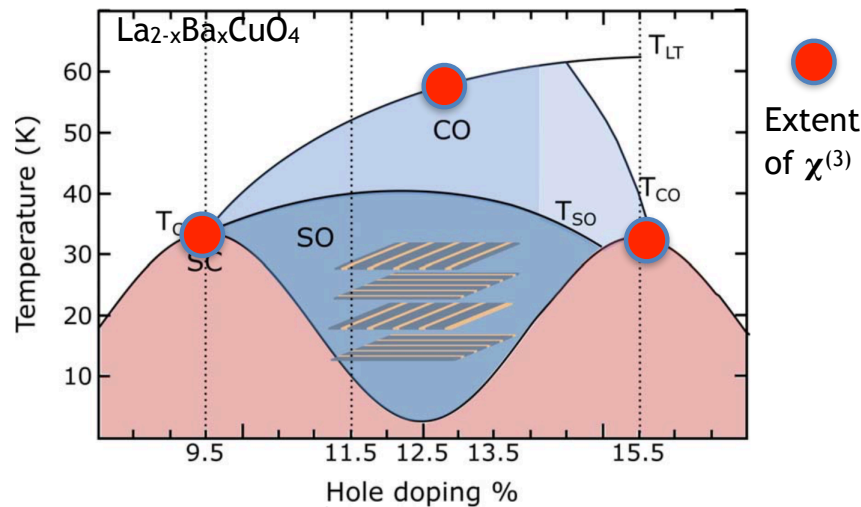
Superconductivity?
Charge Density Waves?
Nonlinearities from strong Correlations?

SUPERCONDUCTIVITY

Probing optically silent superfluid stripes in cuprates

S. Rajasekaran,^{1*} J. Okamoto,² L. Mathey,² M. Fechner,¹ V. Thampy,³
G. D. Gu,³ A. Cavalleri^{1,4*}

not easily validated with linear optics, because the stripe alignment causes interlayer superconducting tunneling to vanish on average. Here we show that this frustration is removed in the nonlinear optical response. A giant **terahertz third harmonic**, characteristic of nonlinear Josephson tunneling, is observed in $\text{La}_{1.885}\text{Ba}_{0.115}\text{CuO}_4$ above the transition temperature $T_c = 13$ kelvin and up to the charge-ordering temperature $T_{co} = 55$ kelvin. We model these results by hypothesizing the presence of a pair density wave condensate, in which nonlinear mixing of optically silent tunneling modes drives



Conclusions

- 2D coherent THz spectroscopy is a new nonlinear $\chi^{(3)}$ technique with vast potential for quantum materials
- Can get unique information about spin liquids (experiments ongoing... theory now only)
- Photon echo observed in a electron glass $\rightarrow$ not continuable to weakly interacting disordered “Anderson insulator”; a “non-Fermi glass”
- Cuprate superconductors show a large regime of normal state $\chi^{(3)}$ nonlinearity; anomalous response unlike that of normal metals