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

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

\[ P_i = \chi_{ijk}^e E_j \]
\[ P_i = \chi_{ijkl}^e E_j E_k E_l \]
\[ M_i = \chi_{ijkl}^m B_j B_k B_l \]
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
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.

\[ E_{NL} = E_{AB} - E_A - E_B \]
2D NMR Spectroscopy

Nuclear Overhauser Effect Spectroscopy (NOESY) spectra of codeine
2D THz Coherent Spectroscopy

Astrella

800 nm, 30 fs
7mJ/pulse
Excite with two pulses. Slide through each other in time. Look at difference signal with both together from each separately.

\[ E_{\text{NL}} = E_{\text{AB}} - E_A - E_B \]
Third order non-linear response

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

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

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

\[ \propto \chi_{ijkk}[(EE)E e^{i(2\omega_j + \omega_k)t} + (EE*)E e^{i(\omega_j - \omega_j + \omega_k)t} + (E*E)E e^{i(-\omega_j + \omega_j + \omega_k)t} + (E*E*)E e^{i(-2\omega_j + \omega_k)t} + (E*E)E* e^{i(\omega_j - \omega_j - \omega_k)t} + (E*E*)E* e^{i(-2\omega_j - \omega_k)t}] + \]

\[ + \chi_{ijjk}[(EE)e^{i(\omega_j + 2\omega_k)t} + (EE*)e^{i(\omega_j + \omega_k)2\omega_k)t} + (E*E)e^{i(\omega_j - \omega_k + \omega_k)t} + (E*E*)e^{i(\omega_j - 2\omega_k)t}] \]

3HG

\[ \Delta P_i \propto \chi_{ijkk}[(EE)e^{i(2\omega_j + \omega_k)t} + (EE*)e^{i(\omega_j - \omega_j + \omega_k)t} + (E*E)e^{i(-\omega_j + \omega_j + \omega_k)t} + (E*E*)e^{i(2\omega_j - \omega_k)t}] + \]

\[ + \chi_{ijjk}[(EE)e^{i(\omega_j + 2\omega_k)t} + (EE*)e^{i(\omega_j + \omega_k)2\omega_k)t} + (E*E)e^{i(\omega_j - \omega_k + \omega_k)t} + (E*E*)e^{i(\omega_j - 2\omega_k)t}] + \text{c.c.} \]

3HG

PP

PP

2Q

NR

NR

R
Example: Magnons in AFM

- YFeO$_3$ : Canted Anti-ferromagnet

Lu, Nelson PRL 2017
Spin systems with fractionalization
Spin singlet formation in a spin liquid

Slide courtesy of L. Balents
Spinon Continuum in 1D KCuF₃

B. Lake et al. (2013)

Mourigal et al. (2013)
Fractionalization: 1D Ising chain in small transverse field

\[ \Delta = 4J(1 - \frac{h}{J}) \]

\[ \Delta = 4J(1 + \frac{h}{J}) \]

*CoNb_2O_6*

*\( T = 3K \)*

Absorption (a.u.)

Frequency (THz)

\( \Delta = 4J(1 - \frac{h}{J}) \)

\( \Delta = 4J(1 + \frac{h}{J}) \)
Purported Spinon Continuum in higher dimension

YbMgGaO$_4$

Evidence for spinon continuum less convincing in higher D.

Broad spectral shapes hard to distinguish from disorder

Shen et al. 2017
\[ H = -J \sum_{n=1}^{L-1} \sigma_n^z \sigma_{n+1}^z + J \sigma_L^z \sigma_1^z - h \sum_n \sigma_n^x \]

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 \( n_c \)

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

\( \xi \sim 13 \text{ nm} \)
\( n_P^{-1/3} \sim 9 \text{ nm} \)
\( r_{\text{Bohr}} \sim 2.5 \text{ nm} \)
\( \varepsilon \sim 14 \) (\( \varepsilon_{\text{Si}} \sim 11.7 \))

\( \xi > n_P^{-1/3} \)

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

Rosenbaum et al. PRL 1983
Ensemble of orbitals localized in real space due to disorder — *Anderson insulator*
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.
At low $\omega$ 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.

$$\sigma = A\omega \left[ \frac{\hbar \omega + e^2}{\varepsilon_1 \langle r_\omega \rangle} \right]$$

Resonant pair approximation

G. Thomas et al.

Helgren, NPA, Gruner, 2002, 2004
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

\[ \frac{x}{x_c} = 39\% \]
\[ \frac{x}{x_c} = 50\% \]
\[ \frac{x}{x_c} = 55\% \]
\[ \frac{x}{x_c} = 62\% \]
\[ \frac{x}{x_c} = 70\% \]
\[ \frac{x}{x_c} = 85\% \]
2D Terahertz spectroscopy on a localized electron glass system

\[ \frac{x}{x_c} = 39\% \]

\[ \frac{1}{T_1} \]

\[ \frac{1}{T_2} \]

\[ T = 5\, K \]

\[ \nu - \nu_c \text{ (THz)} \]

\[ |\text{FFT}| \text{ (norm.)} \]

\[ \text{NR quadrant} \]

\[ \text{R quadrant} \]

\[ Cuts \text{ along the NR quadrant} \]

\[ \frac{1}{T} \]

\[ \frac{1}{T_1} \]

\[ \frac{1}{T_2} \]

\[ P: Si 0.02 \]

\[ \text{Rate (THz)} \]

\[ \text{Doping dependence of } \frac{1}{T_1} \text{ and } \frac{1}{T_2} \]
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”
La$_{2-x}$Cu$_x$O$_4$
Optical response from one intense THz pulse

Non-Linear response from two overlapping intense THz pulses

LSCO \( (T_c = 27.5K) \)

NbN \( (T_c = 15K) \)

Au

Transmission vs. Frequency (THz)

\( E_{NL} \) (kV/cm) vs. Time (ps)

\( \Delta T \) vs. E. Field (KV/cm)

\( E_{NL} \) (a.u.) vs. Frequency (THz)
Temperature dependence of THz nonlinearity spectroscopy contributions

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

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

$\nu_s \propto E$
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

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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 La$_{1.885}$Ba$_{0.115}$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...
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