# Can pulse coherence reveal the low energy charge modes in condensed matter?

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# Why do we care about charge modes?

**21st century grand challenge (apologies for DOE speak):**

Understand the physics of interacting electron systems, in particular the doped-Mott insulator

**We "understand" an interacting electron system when …**

1. We can deduce its ground state (preferably a Slater determinant)
2. We can describe its elementary excitations

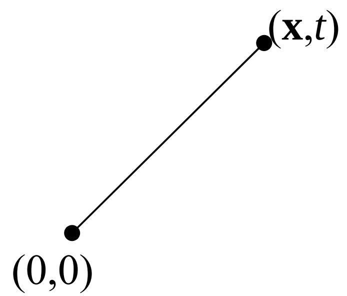
**"Charge modes"** ≡ those excitations that modulate the electron density

**Characterized by dynamic structure factor**,

$S(\mathbf{k},\omega) = \Sigma_n |<n|\rho(\mathbf{k})|0>|^2\, \delta(\omega-\omega_n) = -1/\pi\ \mathrm{Im}[\, \chi(\mathbf{k},\omega)\, ]$

Measured directly by inelastic x-ray or electron scattering

$$\chi(\mathbf{x},t) = -i < 0|\, \hat{n}(\mathbf{x},t)\, \hat{n}(0,0)\, |0> \theta(t)$$



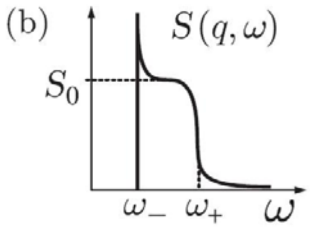
# Why do we care about charge modes?

**A Classic Example:**

Interacting electron gas in RPA (Pines & Nozieres, 1952)

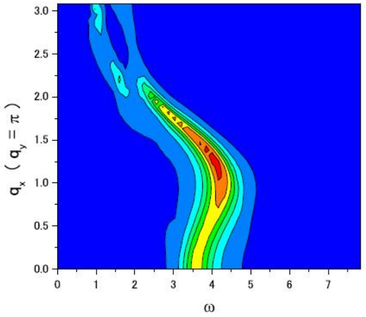
S(**k**,ω
)

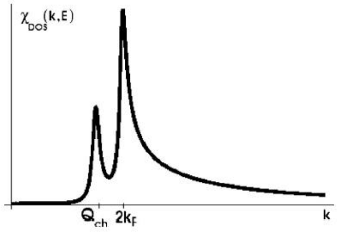
**Some Recent Examples:**




Luttinger liquid with nonlinear dispersion
Pustilnik, *PRL*, **96**,196405 (2006)

Amplitude modes in a d-wave superconductor,
Lee & Nagaosa, *PRB*, **68**, 24516 (2003)



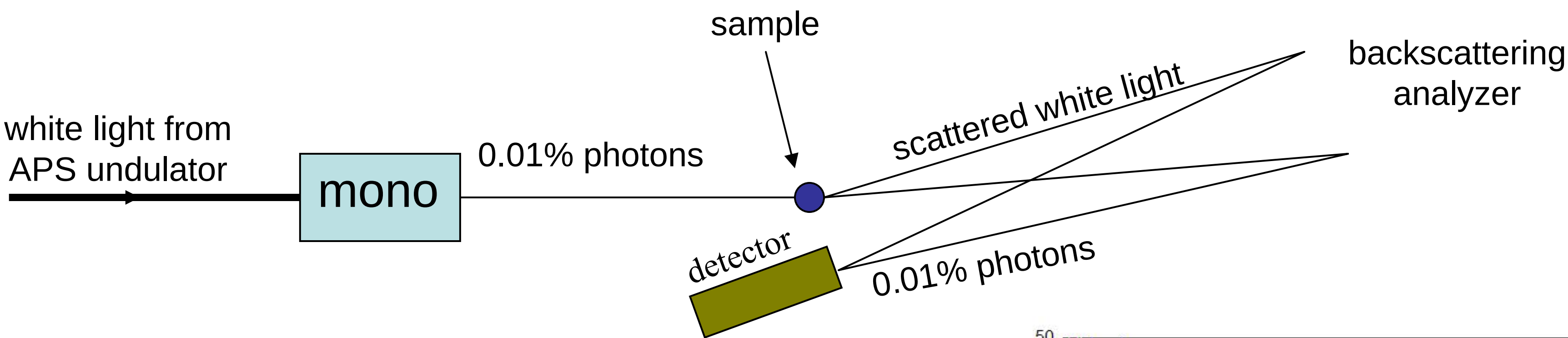




Stripe theory of HTSC
Kivelson, Bindloss, Fradkin, et. al., *RMP*, **75**, 1201 (2004)

# Inelastic x-ray scattering – why doesn't it work?!

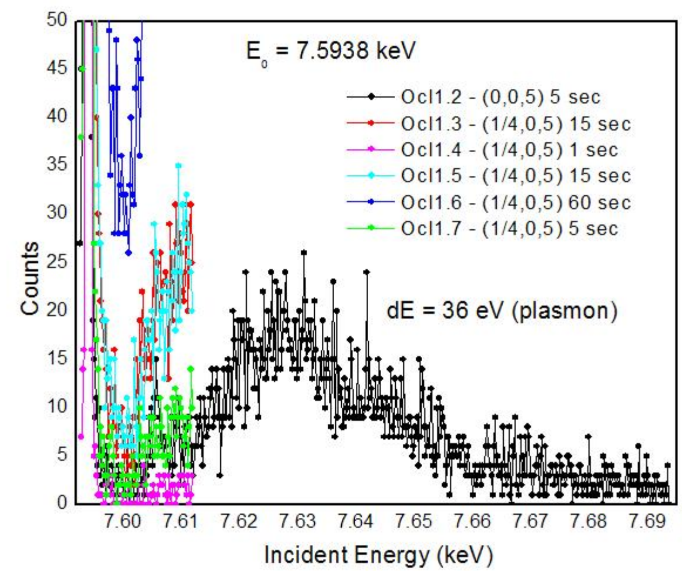
IXS measures $S(\mathbf{k},\omega)$, i.e. the charge response. Why doesn't it work?

1. XRD is coherent, scales like $N^2$
2. IXS incoherent, scales like $N_{valence}$ ~ 0.1 e << N
3. Mono and analyzer both throw away 99.9999% of the photons

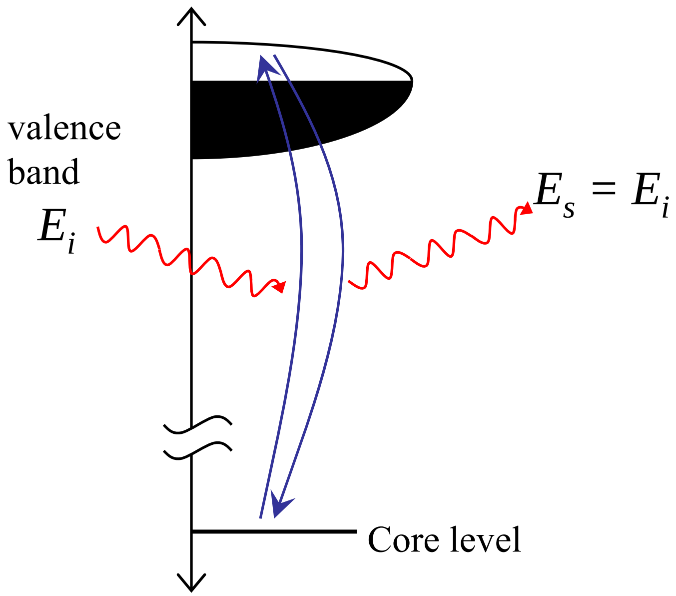


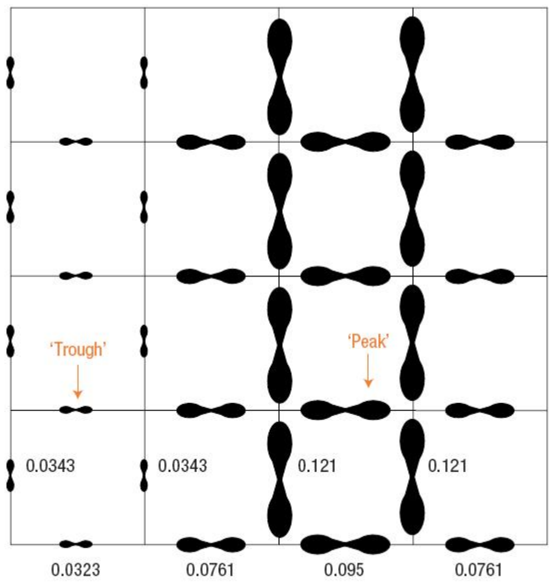

**Upshot: Signal is weak and sitting on tail of elastic, which is stronger by $N^2/N_{valence}$**

$Ca_{2-x}Na_xCuO_2Cl_2$



# Resonance techniques - diffraction



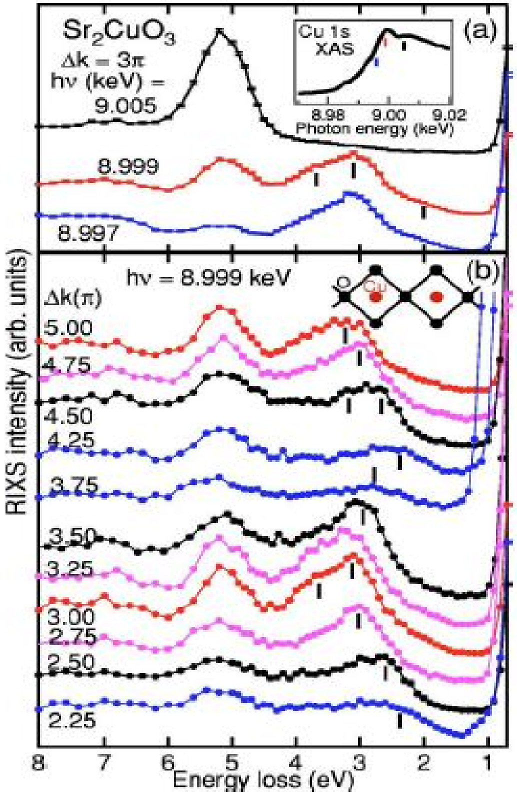

Charge amplitude of static stripes
Abbamonte, Rusydi, et. al., *Nature Physics*, **1**, 155 (2005)

$\Delta v$ = 0.063 holes
$A$ = 0.59 holes

Wigner crystal melting, Rusydi, et. al, *PRL* (2006)
Wigner crystallization in ladders, Abbamonte, et. al., *Nature*, **431**, 1078 (2004)
Charge disproportionation in PCMO, Thomas, et. al. *PRL*, **92**, 237204 (2004)
Depletion zone at an interface, Abbamonte, et. al., *Science*, **297**, 581 (2002)

Are these correlations present dynamically in SC state?

# Resonance techniques – inelastic scattering (RIXS)

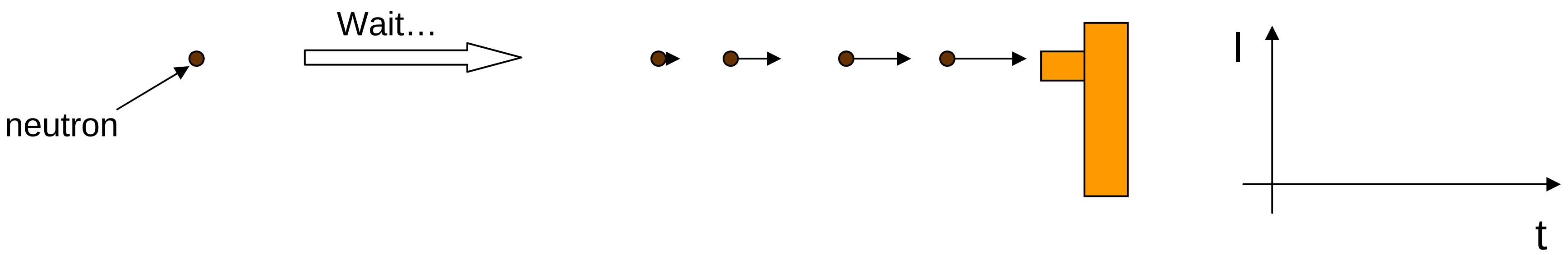



S. Suga, et. al., *PRB*, **72**, 81101(R) (2005)

We're doing chemistry.  Useful, but not good enough.

# Work in time domain?

**Neutron "time-of-flight" method**:




- Uses all neutrons in the bunch (no monochromator)
- No a-priori limit on time resolution
- Does not work for photons because they all travel at c. (except maybe near an edge)

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**Q: How can it be that the frequency structure changes but not the time structure?**

$$|\mathbf{k}\rangle \rightarrow \exp(i\omega_n t)\,|\mathbf{k}\rangle$$

No change in time trace, $E^2(t)$.

# Phase space reformulation

System in its ground state: $|0\rangle$

System in its ground state, plus a photon: $a^{\dagger}_{\mathbf{k}}\,|0\rangle$

Single photon wave packet: $|\psi\rangle = \Sigma_{\mathbf{k}}\, A(\mathbf{k})\, a^{\dagger}_{\mathbf{k}}|0\rangle$
Satisfies $\Delta E \Delta t = \hbar/2$

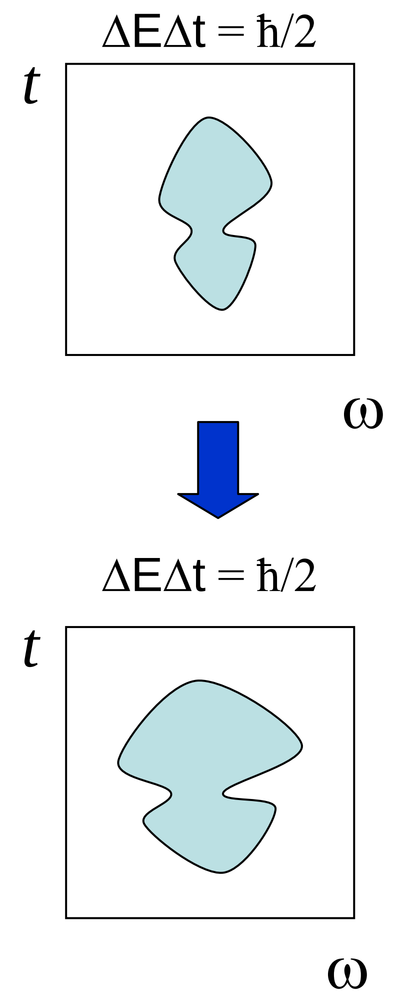
Many photons: $|\psi\rangle = [\Sigma_{\mathbf{k}}\, A(\mathbf{k}_1) a^{\dagger}_{\mathbf{k1}}]\, [\Sigma_{\mathbf{k}}\, A(\mathbf{k}_1) a^{\dagger}_{\mathbf{k1}}] \ldots |0\rangle$
Still satisfies $\Delta E \Delta t = \hbar/2$

Inelastic x-ray scattering:
System undergoes transition: $|0\rangle \rightarrow |n\rangle$
Probability of doing this: $|\langle n|\, \rho(\mathbf{k})\, |0\rangle|^2$
Photon undergoes frequency shift: $|\psi\rangle \rightarrow \exp(i\omega_n t)\, |\psi\rangle$
No change in time trace, $E^2(t)$
BUT … incoherent sum $\Rightarrow$ increase in phase space area

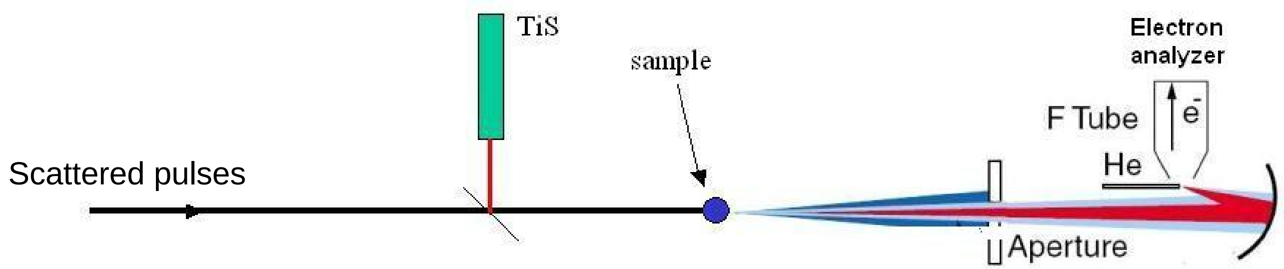
Scattered pulse is only partially coherent.
Contains noise.

Phase space:

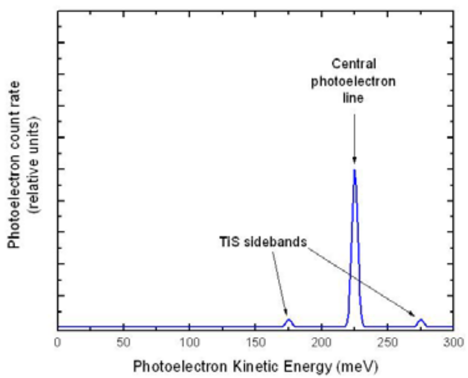
$E(\omega)\cdot E(t)$

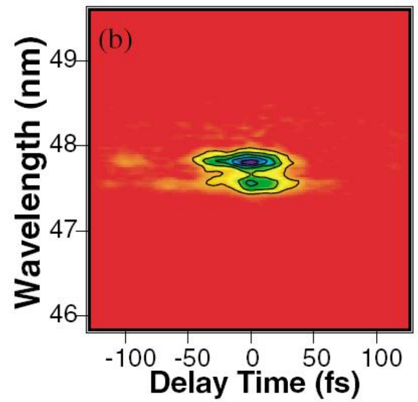


# Possible route: X-FROG




Adapted from Sekikawa, *PRL*, **91**, 103902 (2003)




X-FROG at 25 eV

Tune time delay. Complete phase space reconstruction via simple matrix inversion, provided TiS pulse shape is known

- Entire scattered pulse used at once. More efficient than monochromator.
- Resolution determined by delay line. No intrinsic limit. $\Delta E = 2\pi/\text{rep rate} \sim 3.3$ peV @ 10Hz
- Needs transform-limited pulses
- X-ray analogue to neutron time-of-flight or spin echo

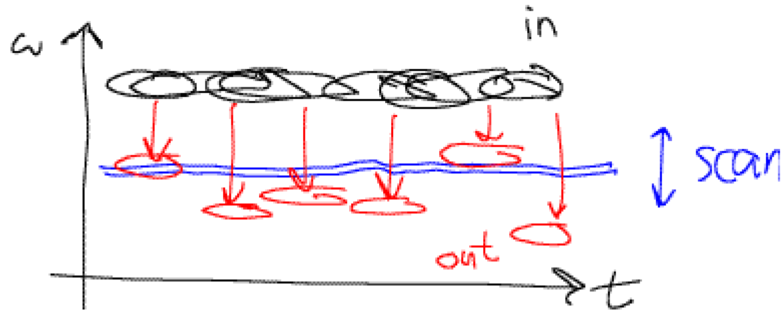
# Conclusions

- IXS is really measurement of violation of Liouville's theorem
- Coherent pulses $\Rightarrow$ more ways to do this than with monochromators
- X-FROG is one possible alternative

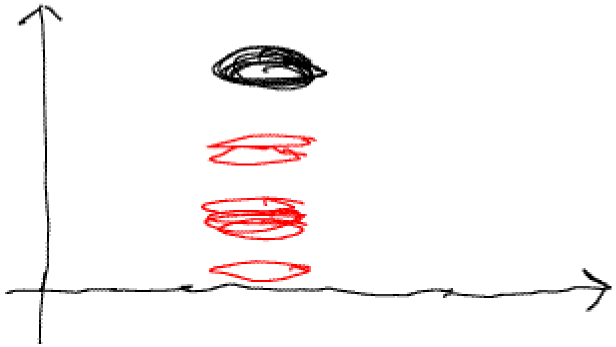
**Detectors**: Windowless APDs

## How to probe decoherence?

Incoherent pulses - “regular IXS”




Coherent pulses



Can detect instead through noise pattern in E(t)