Searching for pairing interactions with coherent charge fluctuations spectroscopy

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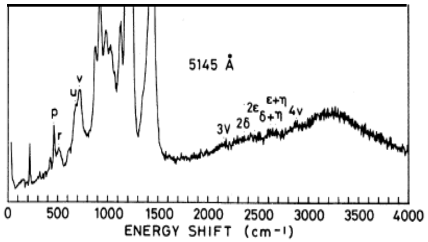
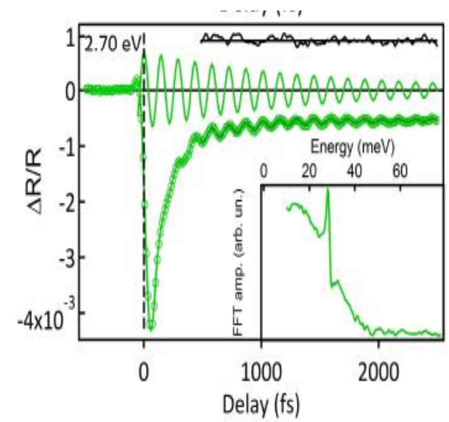
Consiglio Nazionale delle Ricerche

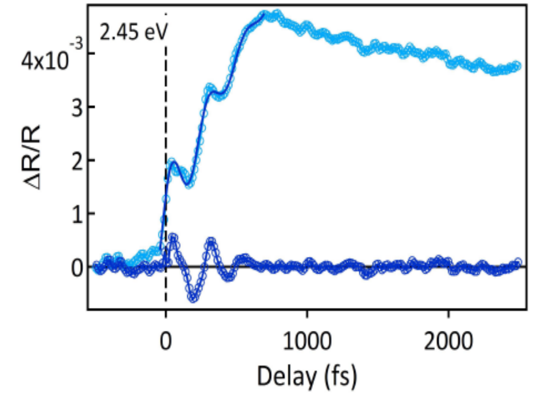
Outline

- Raman scattering

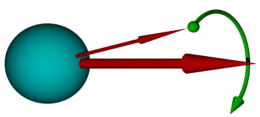




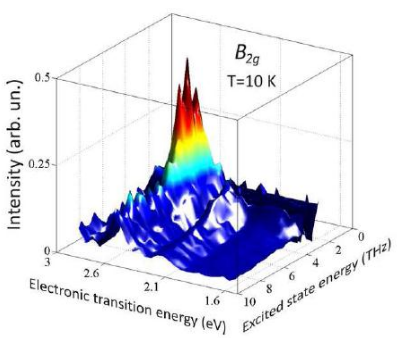
- Coherent Lattice Fluctuation Spectroscopy
- Coherent Charge Fluctuation Spectroscopy



- Coherent oscillations in a superconductor
- NMR in charge space

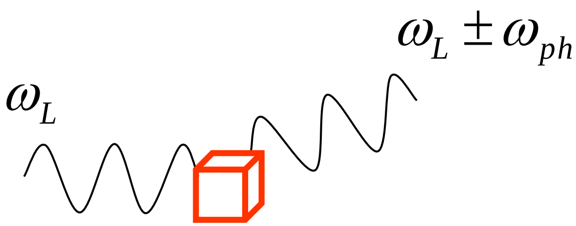


- Condensate coupling with a high-energy mode



Raman Scattering

$$\mathbf{E} = \mathbf{E}_0 e^{-i\omega_L t}$$



$$\mathbf{P} = \boldsymbol{\chi}.\mathbf{E} \qquad \varepsilon = 1 - 4\pi\boldsymbol{\chi}$$

$$\boldsymbol{\chi} = \frac{\sigma}{i\omega}$$

Sir Chandrasekhara Venkata Raman

$$\boldsymbol{\chi} = \boldsymbol{\chi}_0 + \frac{\partial \boldsymbol{\chi}}{\partial \xi}\xi + \frac{\partial \boldsymbol{\chi}}{\partial \xi^*}\xi^*$$

Raman tensor

$$\mathbf{P} = \left(\boldsymbol{\chi}_0 e^{-i\omega_L t} + \frac{\partial \boldsymbol{\chi}}{\partial \xi}\xi e^{-i(\omega_L + \omega_{ph})t} + \frac{\partial \boldsymbol{\chi}}{\partial \xi^*}\xi^* e^{-i(\omega_L - \omega_{ph})t} \right)\mathbf{E}_0$$

Spontaneous Raman Scattering

$$H = H_{ph} - \frac{1}{2}\mathbf{E}(t).\frac{\partial \chi(\omega_L)}{\partial \xi}.\mathbf{E}(t)\hat{\xi}$$

$$\hat{\boldsymbol{\rho}} = \frac{\partial \boldsymbol{\chi}}{\partial \xi}(\omega_L)\hat{\xi}$$

$$\frac{d\sigma}{d\Omega d\omega} = \frac{\omega_s^4 V^2}{(4\pi)^2 c^4}\sum_{\nu} |\langle 0|\hat{e}_s.\hat{\rho}.\hat{e}_l|\nu\rangle|^2 \delta(\omega - \omega_\nu)$$

Sugai PRB ‘89

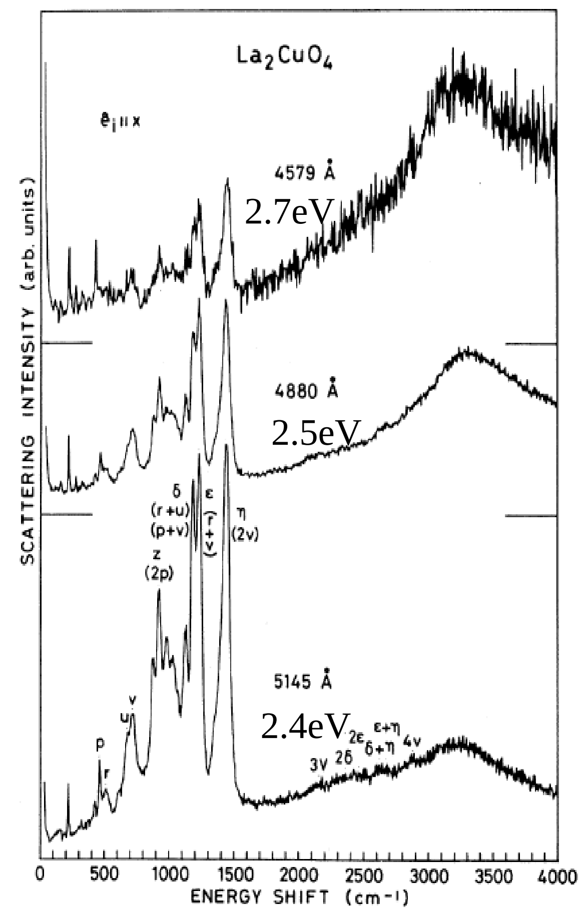


FIG. 10. Incident wavelength dependence of the Raman spectra in La_2CuO_4 at 30 K.

Impulsive Stimulated Raman Scattering and Coherent Lattice Fluctuation Spectroscopy

Merlin ssc 1997, Stevens, Kuhl, Merlin PRB 2002

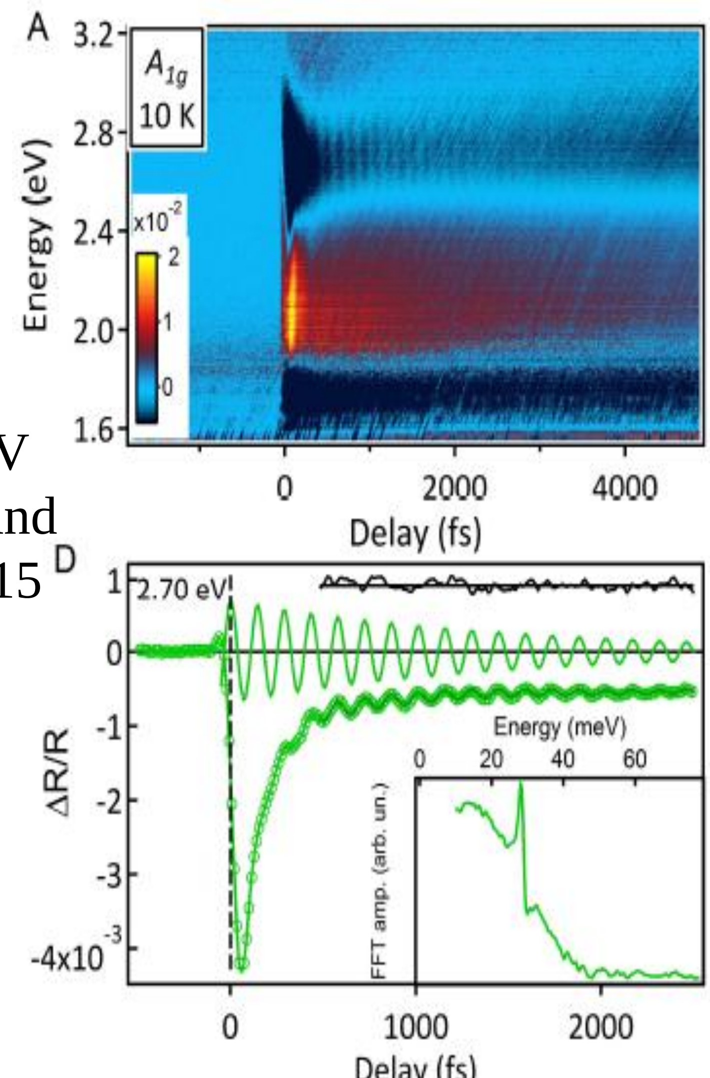
$$H = H_{ph} - \frac{1}{2}\mathbf{E}(t).\frac{\partial\chi(\omega_L)}{\partial\xi}.\mathbf{E}(t)\hat{\xi} = H_{ph} - F(t)\hat{\xi}$$

$$H_{ph} = \frac{1}{2}\Pi^2 + \frac{1}{2}\omega_{ph}^2\xi^2$$

pump || [100] 1.55eV
probe || [001] broad band
La2-x SrxCuO4 x=0.15

$$\ddot{\xi} + \omega_{ph}^2\xi = F(t)$$

$$\xi(t) = \int_{-\infty}^{t} dt' \frac{\sin[\omega_{ph}(t-t')]}{\omega_{ph}} F(t)$$



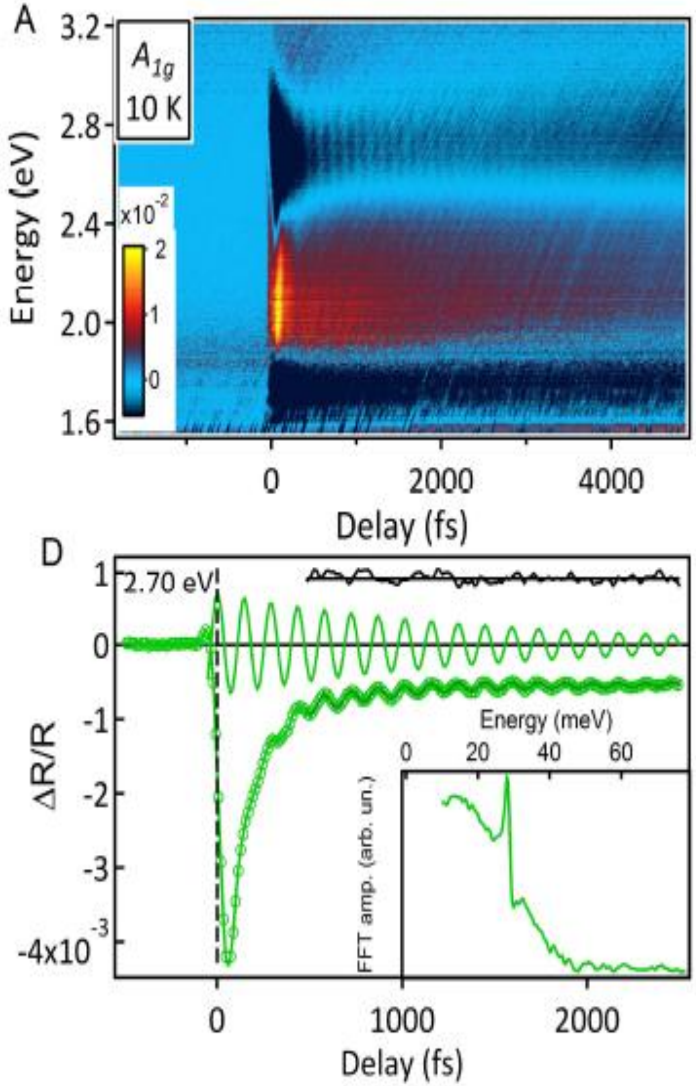
Detection of Excitations

$$\xi(t) = \int_{-\infty}^{t} dt' \frac{\sin[\omega_{ph}(t-t')]}{\omega_{ph}} F(t)$$

$$\boldsymbol{\varepsilon} = 1 - 4\pi\boldsymbol{\chi}$$

$$\delta\epsilon_{\mu\nu}(t) = -4\pi \frac{d\chi_{\mu\nu}}{d\xi}\xi(t)$$

pump || [100]
probe || [001]



Swiss Knife Matrix Element

$$\frac{d\chi_{\mu\nu}}{d\xi} =$$

Spontaneous Raman scattering

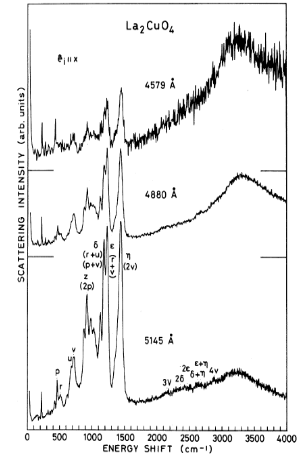
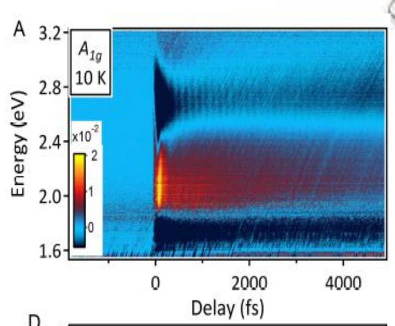


FIG. 10. Incident wavelength dependence of the Raman spectra in La$_2$CuO$_4$ at 30 K.

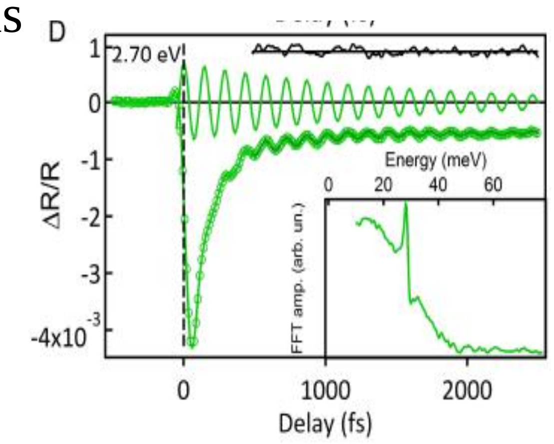
Coherent generation of excitations

Coherent control of excitations

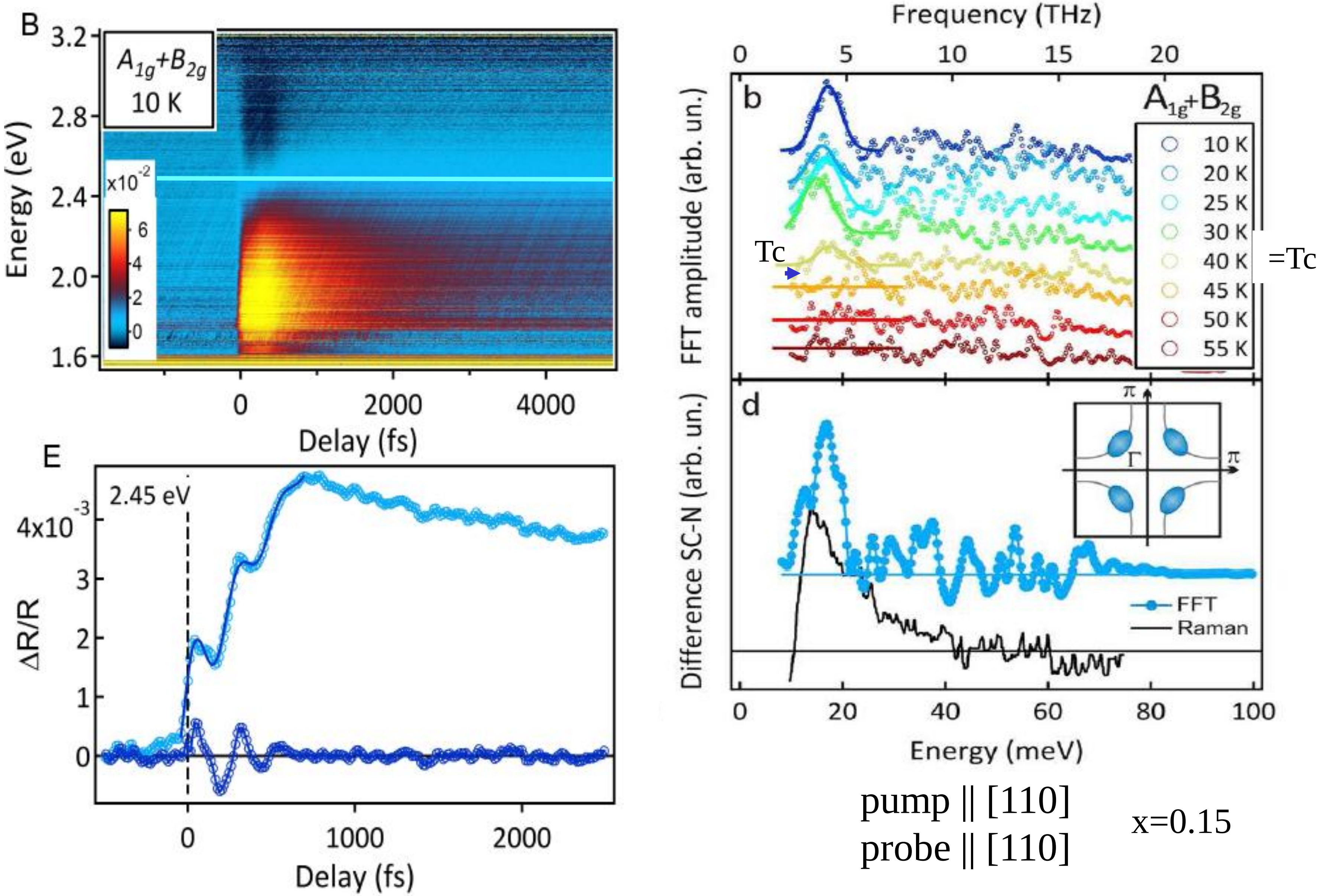
Detection of excitations



Coherent Fluctuation Spectroscopy



A new coherent excitation



pump || [110]
probe || [110]

x=0.15

Coherent generation of excitations

Phonons

$$H = H_{ph} - \frac{1}{2}\mathbf{E}(t).\frac{\partial\chi(\omega_L)}{\partial\xi}.\mathbf{E}(t)\hat{\xi}$$

$$= H_{ph} - F(t)\hat{\xi}$$

$$\xi(t) = \int_{-\infty}^{t} dt' \frac{\sin[\omega_{ph}(t-t')]}{\omega_{ph}} F(t)$$

Charge Fluctuations

$$H = H_{BCS} - \frac{1}{2}\mathbf{E}(t).\frac{\partial\chi(\omega_L)}{\partial N_X}.\mathbf{E}(t)\hat{N}_X$$

$$= H_{BCS} + v_X(t)\hat{N}_X$$

$$\delta N_X(t) =$$

$$-i\int_{-\infty}^{t} dt' \langle[\hat{N}_X(t), \hat{N}_X(t')]\rangle v_X(t)$$

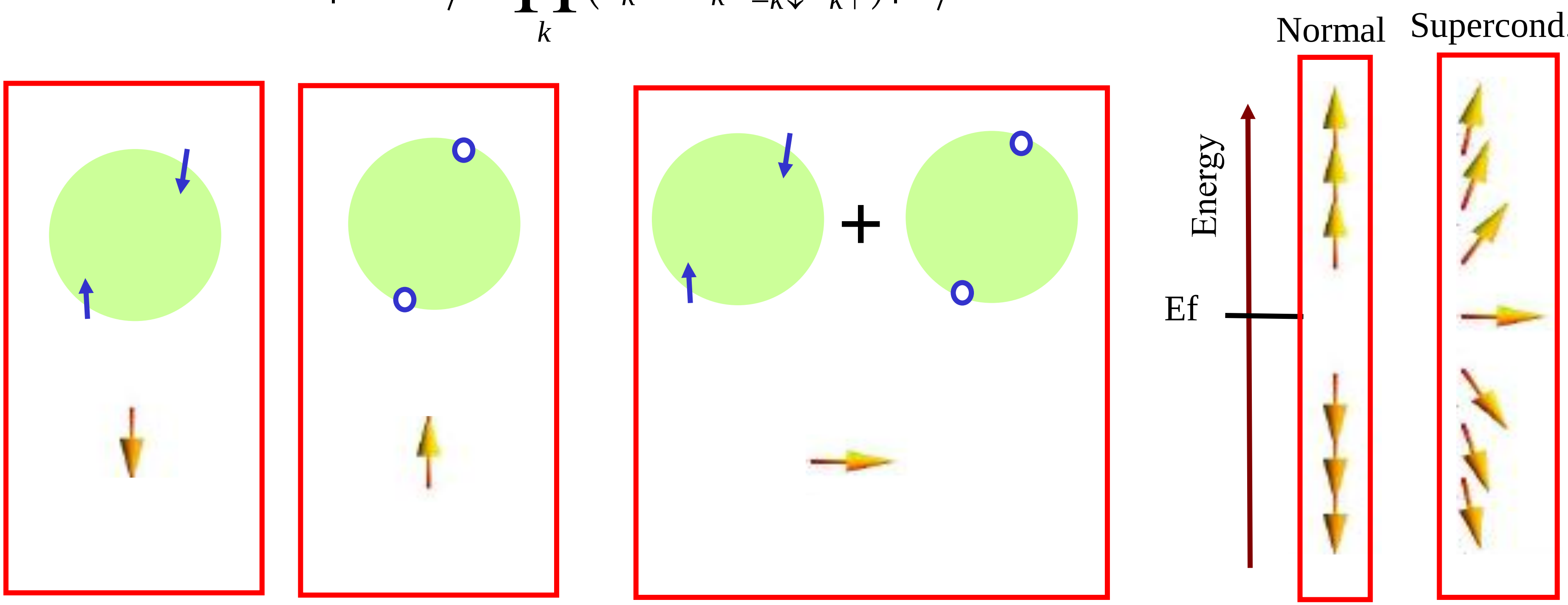
Magnetism and Superconductivity

Anderson Phys. Rev 1958

$$H_{BCS} = \sum_k \xi_k c^\dagger_{k\sigma} c_{k\sigma} - \sum_k (\Delta^*_k c^\dagger_{-k\downarrow} c^\dagger_{k\uparrow} + h.c.)$$

$$\sigma^x_{\mathbf{k}} = (c_{\mathbf{k}\uparrow} c_{-\mathbf{k}\downarrow} + h.c.),\ i\sigma^y_{\mathbf{k}} = (c_{\mathbf{k}\uparrow} c_{-\mathbf{k}\downarrow} - h.c.),\ \sigma^z_{\mathbf{k}} = 1 - n_{\mathbf{k}\uparrow} - n_{-\mathbf{k}\downarrow}$$

$$| BCS \rangle = \prod_k \left(u_k + v_k c^+_{-k\downarrow} c^+_{k\uparrow} \right) | 0 \rangle$$



Magnetism and Superconductivity

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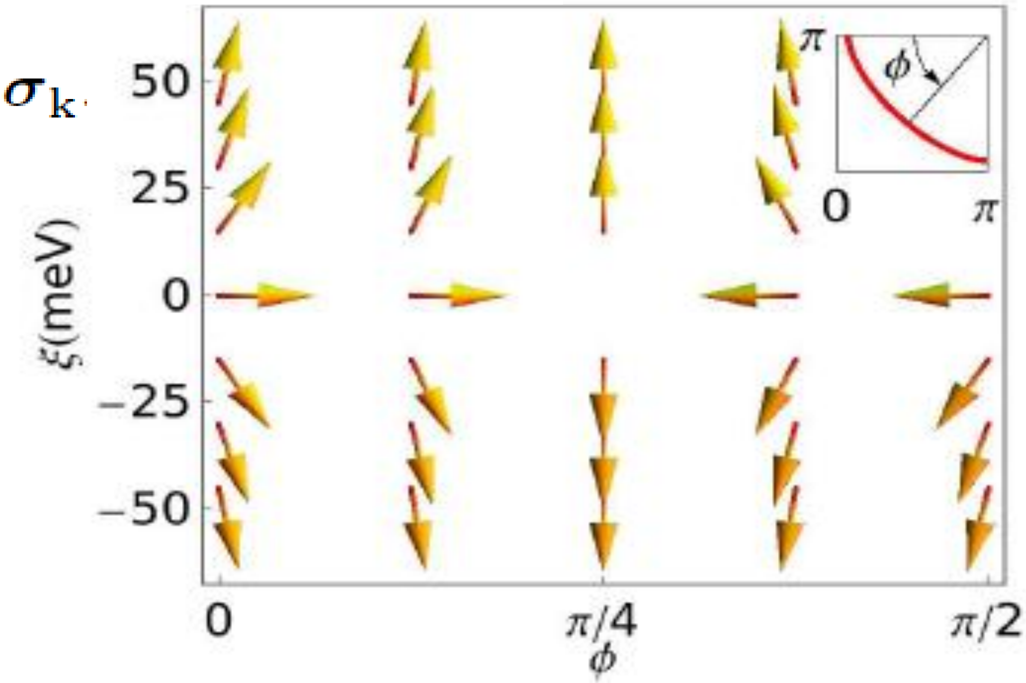
$$H_{BCS} = \sum_k \xi_k c^\dagger_{k\sigma} c_{k\sigma} - \sum_k (\Delta^*_k c^\dagger_{-k\downarrow} c^\dagger_{k\uparrow} + h.c.)$$

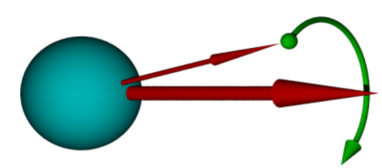
$$H_{BCS} = -\sum_{\mathbf{k}} [\mathbf{b}^0_{\mathbf{k}} + \delta \mathbf{b}_{\mathbf{k}}(t)].\boldsymbol{\sigma}_{\mathbf{k}}.$$

$$\mathbf{b}^0_{\mathbf{k}} = (\Delta_{\mathbf{k}}, 0, \xi_{\mathbf{k}})$$

$$\delta \mathbf{b}_{\mathbf{k}}(t) = (0, 0, v^X_{\mathbf{k}}(t))$$

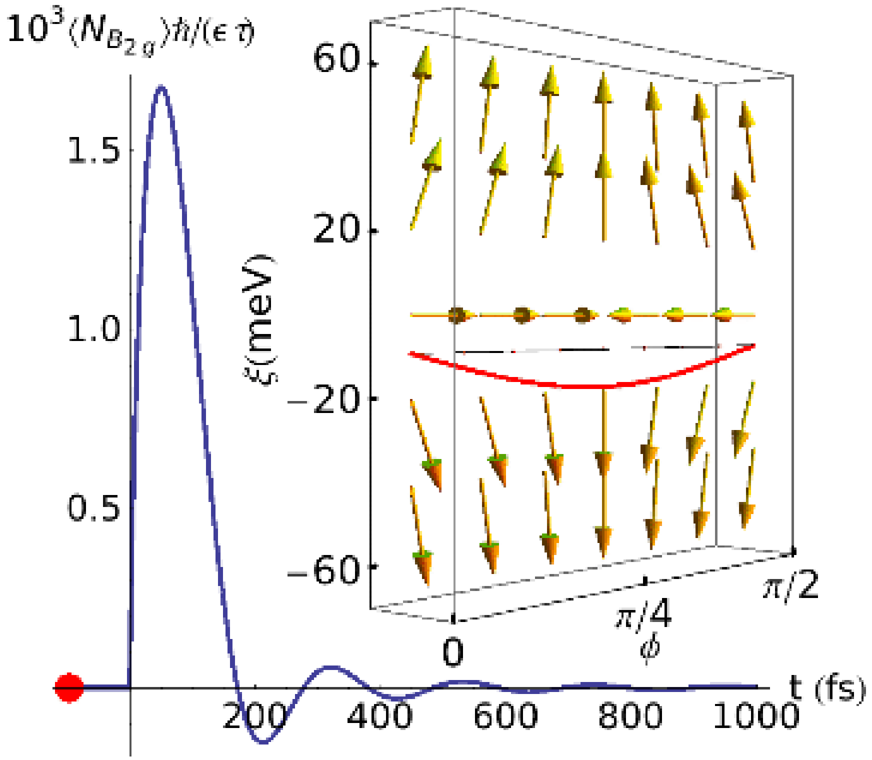


$$\hbar \frac{\partial \boldsymbol{\sigma}_{\mathbf{k}}}{\partial t} = -2[\mathbf{b}^0_{\mathbf{k}} + \delta \mathbf{b}_{\mathbf{k}}(t)] \times \boldsymbol{\sigma}_{\mathbf{k}}.$$ NMR like !



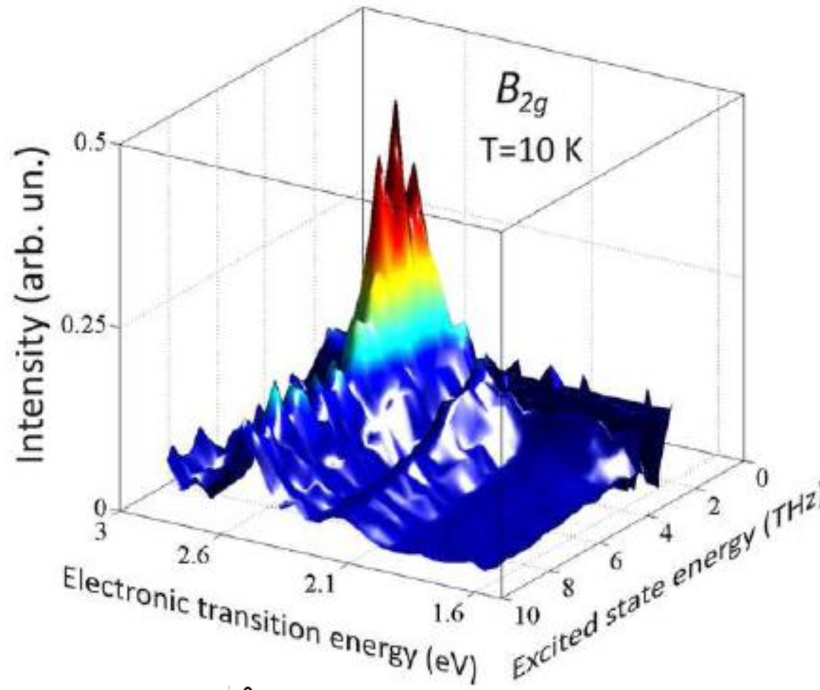
Larmor at Ef $\omega_k = 2\Delta_k$

NMR in Charge Space

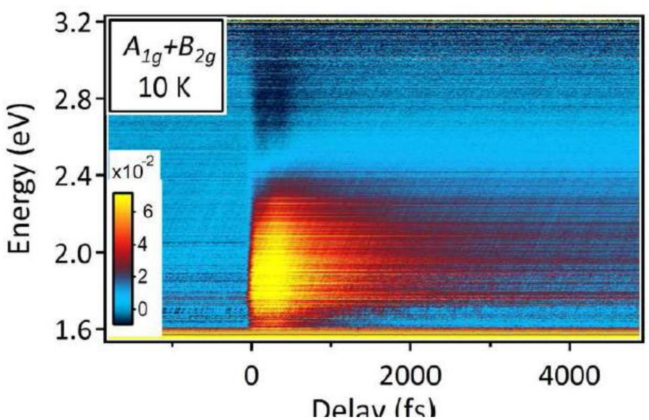


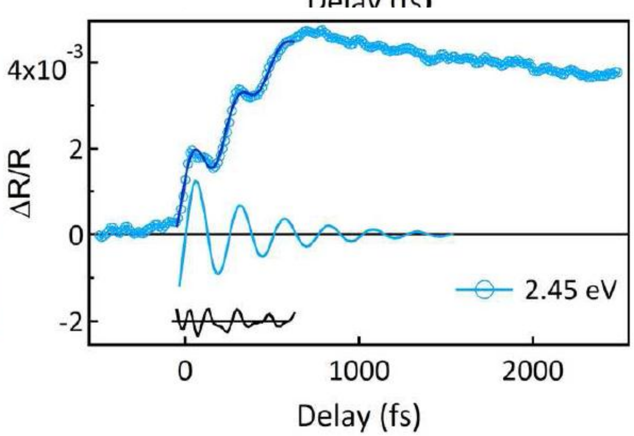
Coherent Charge Fluctuation Spectroscopy

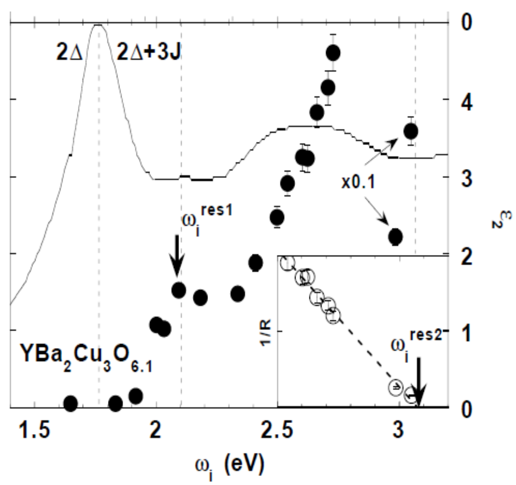
$$\delta\epsilon(\omega, t) = -4\pi \sum_X \frac{\partial\chi}{\partial N_X}(\omega)\langle N_X\rangle(t)$$

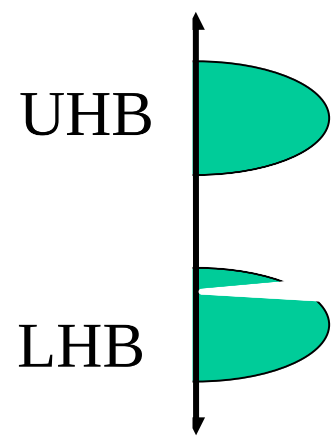


Very specific!





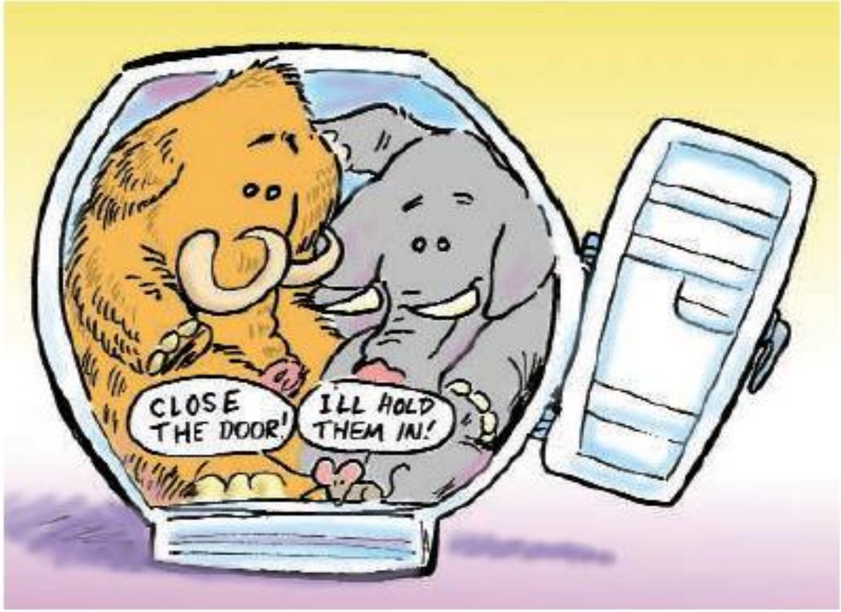




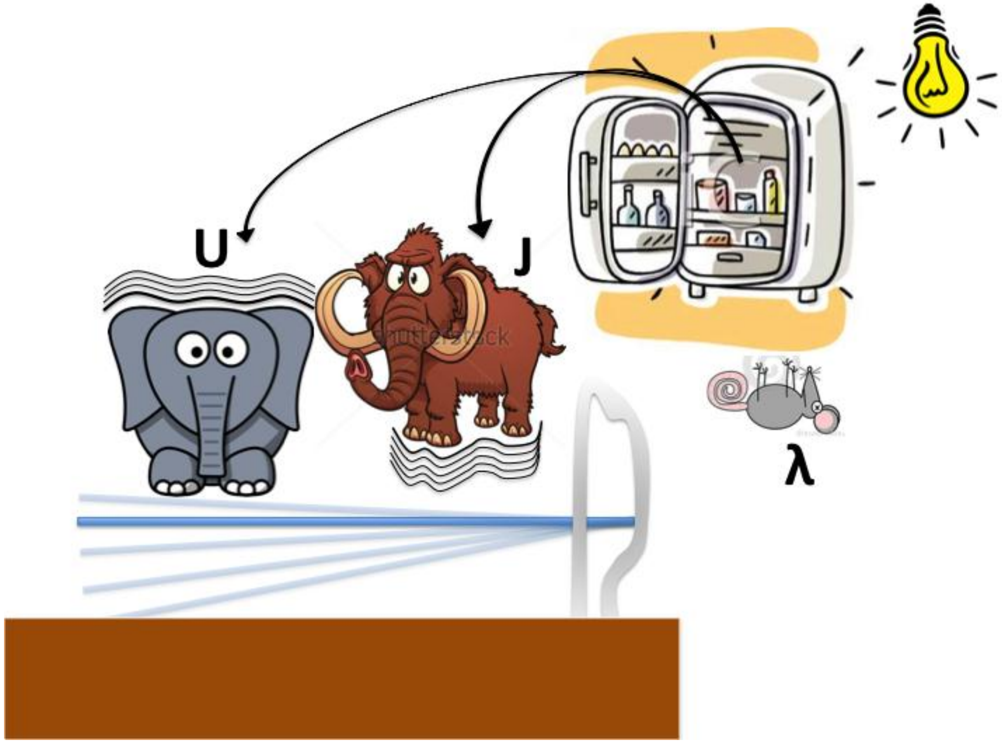
Glue Debate

Anderson, Scalapino, Science 2007

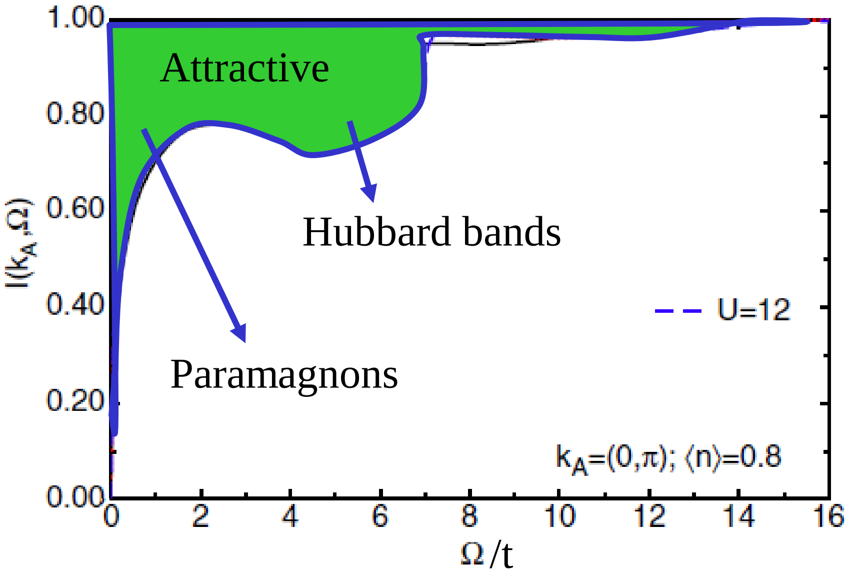
Scalapino: Numerics support a retarded interaction scenario.
Paramagnons are the glue.
Anderson: RVB is the true. Do not search for phonons, magnons, etc.
There is no need to be a glue-sniffer.



"We have a mammoth and an elephant in our refrigerator—
do we care much if there is also a mouse?"



Glue debate



~20% of the attraction from coupling to high energy states

Maier, Poilblanc, D. J. Scalapino, PRL 2008

Conclusions

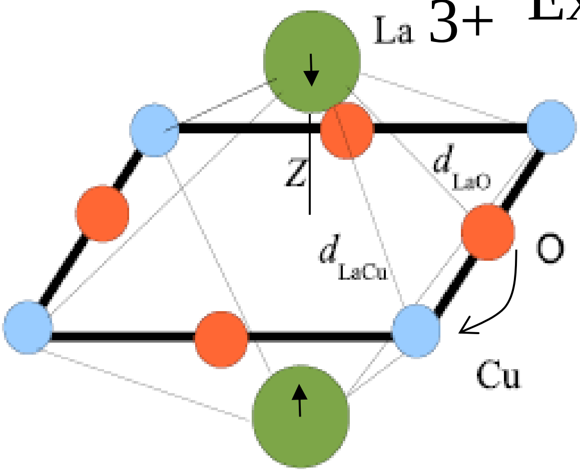
- Superconducting charge fluctuations generated and detected by light pulses for the first time.
- Strong analogy with NMR opens the possibility of coherent control of the superconducting wave function.
- Raman profiles carry precious information on the coupling between low energy excitations and high energy excitations.
- Enables Coherent Charge Fluctuation Spectroscopy, a new technique that allows to answer the question: Which excitations are coupled to the superconducting quasiparticles?. High specificity like Isotope effect.
- In our system: Excitations at the scale of the Hubbard U are coupled to low energy charge fluctuations. Signature of Mottness in the superconducting wave function.

Ref: Mansart et al. PNAS **110**, 4539 (2013).

Lorenzana et al. EPJ ST, **222**, 1223 (2013).

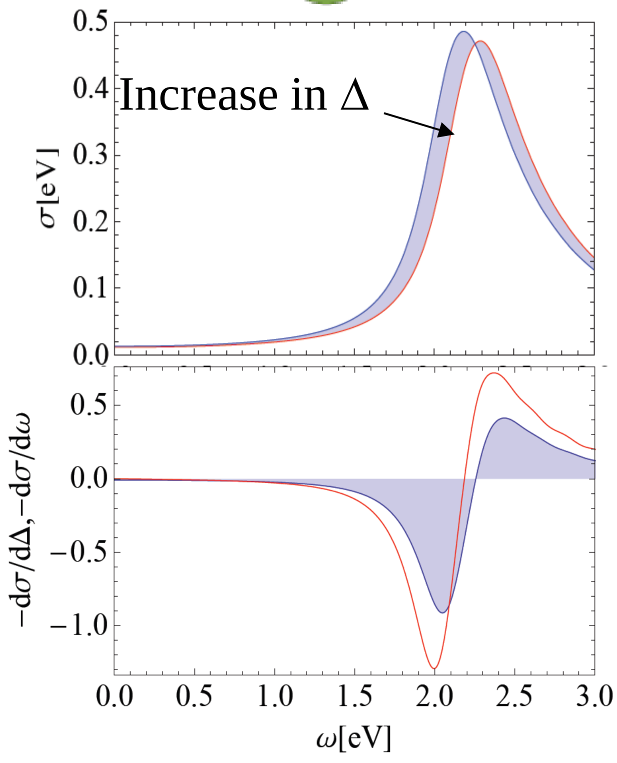
Raman Scattering

Example: The A1g La Phonon



$$\Delta = \Delta_0 + Ze^2\left(\frac{1}{d_{LaO}} - \frac{1}{d_{LaCu}}\right)$$

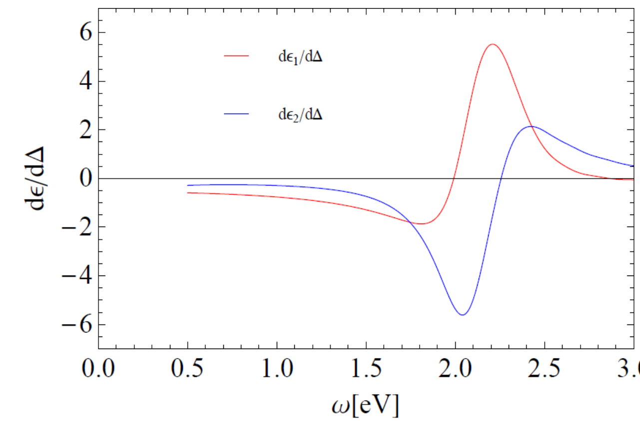
$$\sqrt{M_{La}}\mathbf{u}_{La} = \mathbf{e}_{La}\xi \exp(-i\omega t) + \mathbf{e}^*_{La}\xi^* \exp(i\omega t)$$

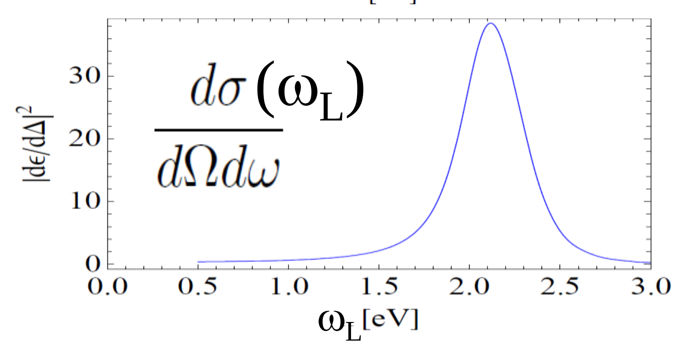


el-ph

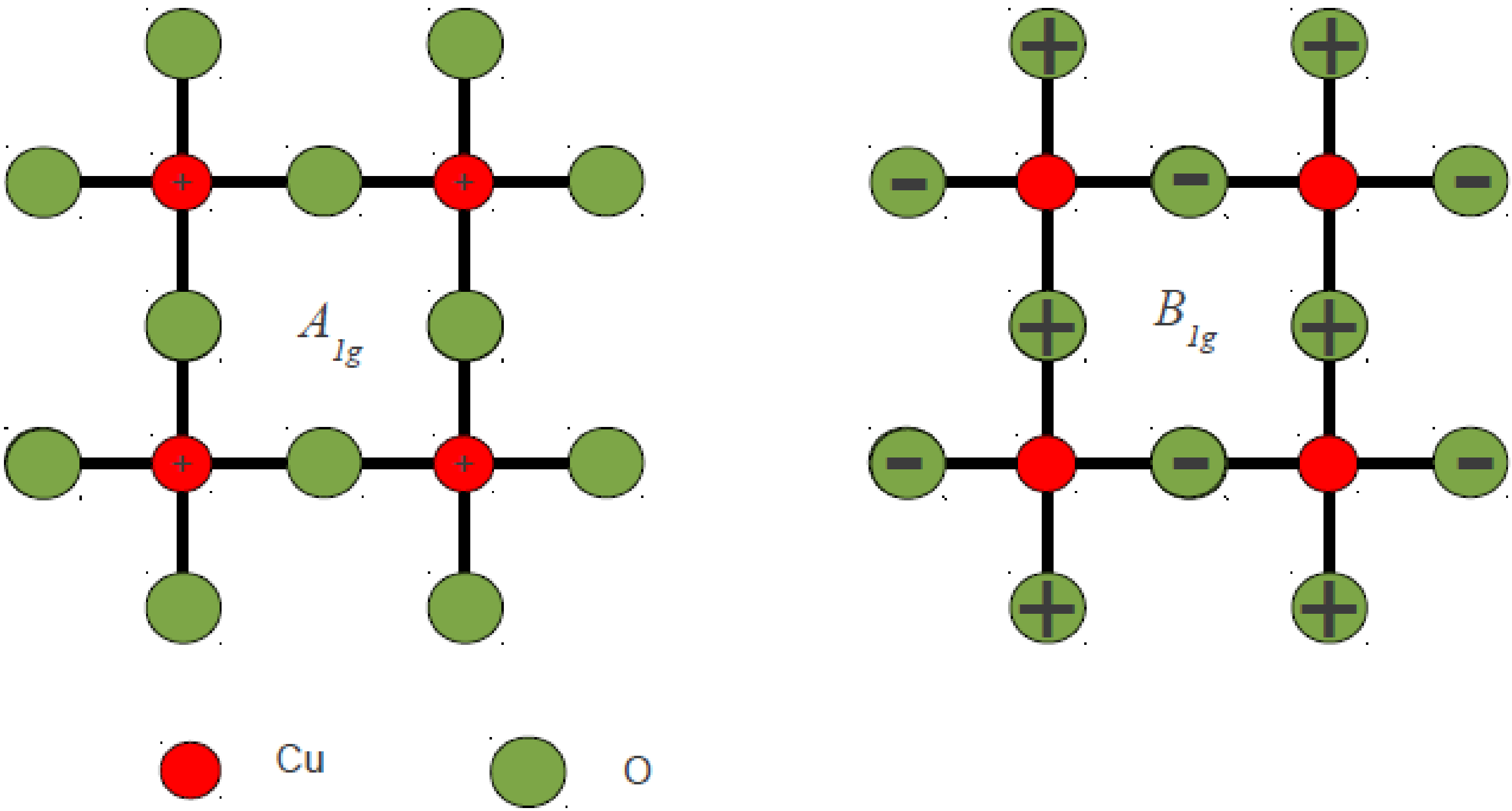
$$\frac{d\chi}{d\xi} = \frac{d\chi}{d\Delta}\frac{d\Delta}{dz}\frac{dz}{d\xi}$$

$$\frac{d\chi}{d\Delta} \approx -\frac{d\chi}{d\omega}$$

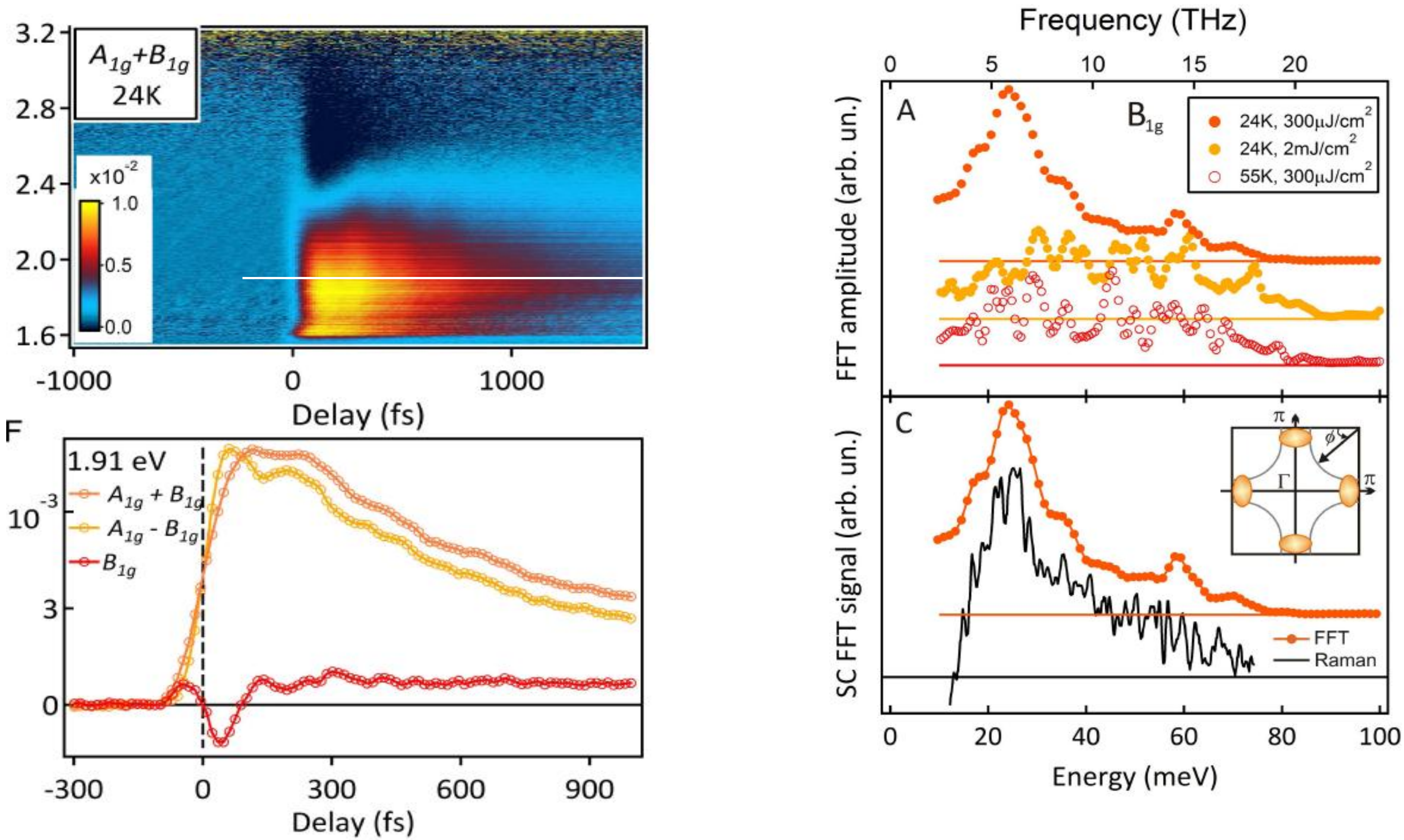




Polarization Analysis

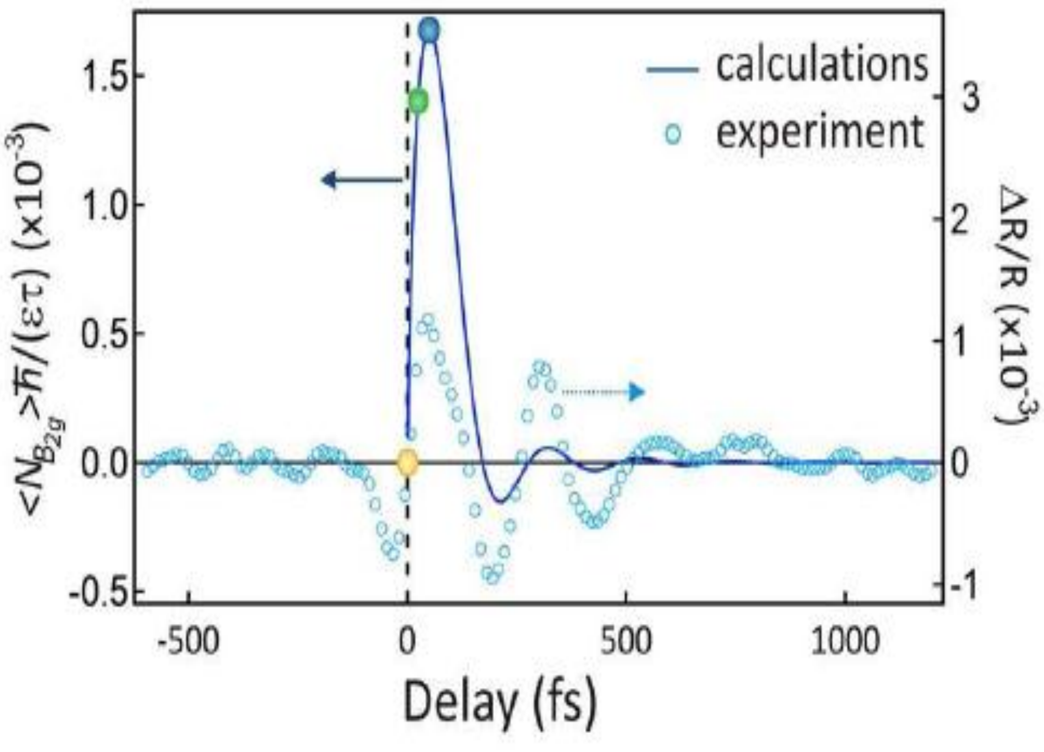


B1g Symmetry and Fluency Dependence



A1g+B1g pump || [100] probe || [100]
A1g-B1g pump || [100] probe || [010]

NMR in Charge Space

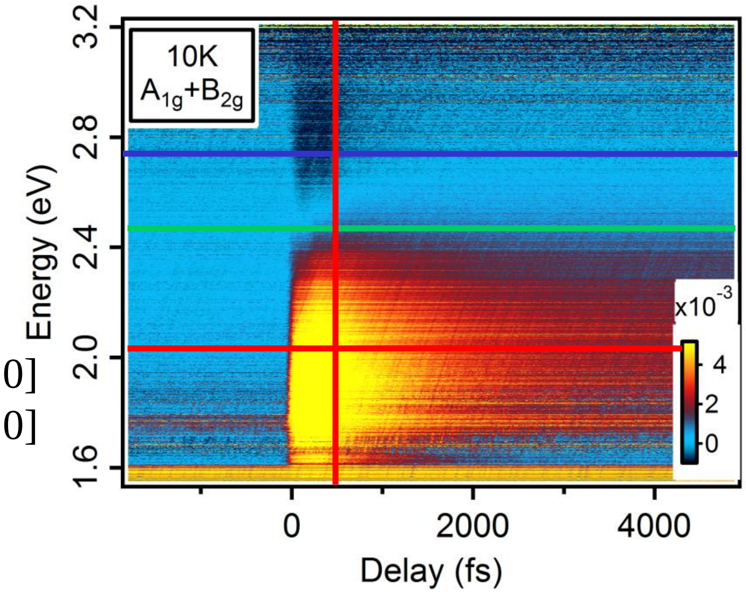


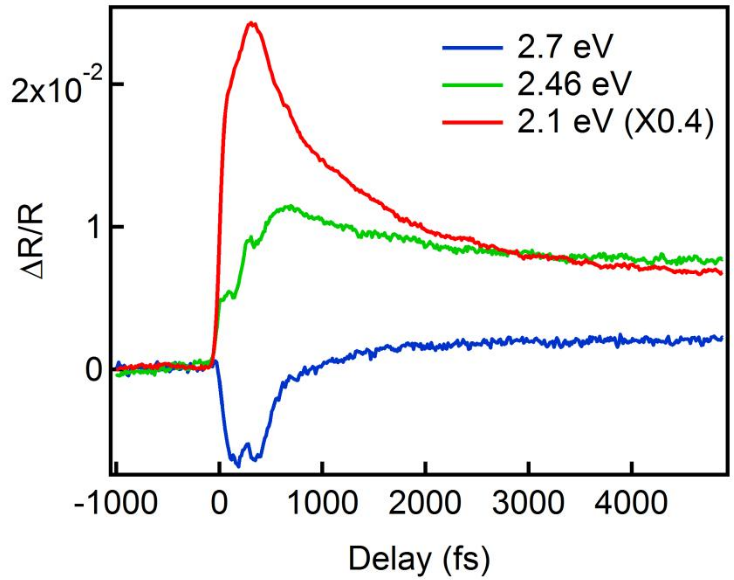
← T_2^* →

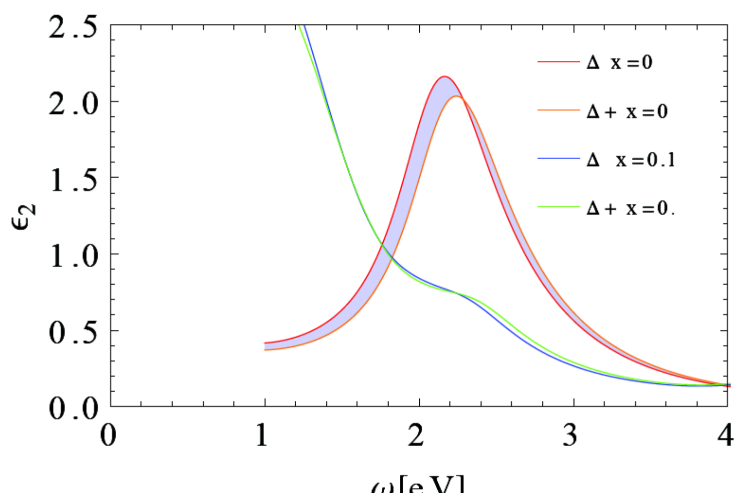
Raman profile as a fingerprint of excitations

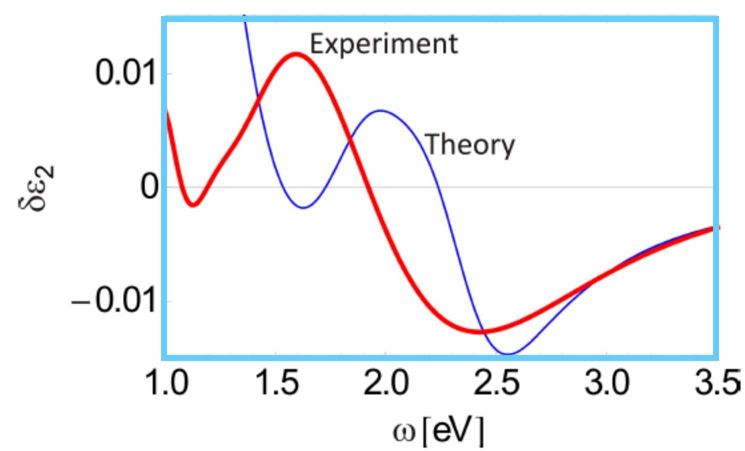
$$\delta\epsilon_{xx}(\omega,t) = -4\pi\frac{\partial\chi_{xx}}{\partial n_{CT}}(\omega)\delta n_{CT}(t)$$

pump || [110]
probe || [110]
X=0.15









Real time Raman vs Frequency Domain

- Phase sensitive information
- Raman profile in one shot
- Coherent control of excitations

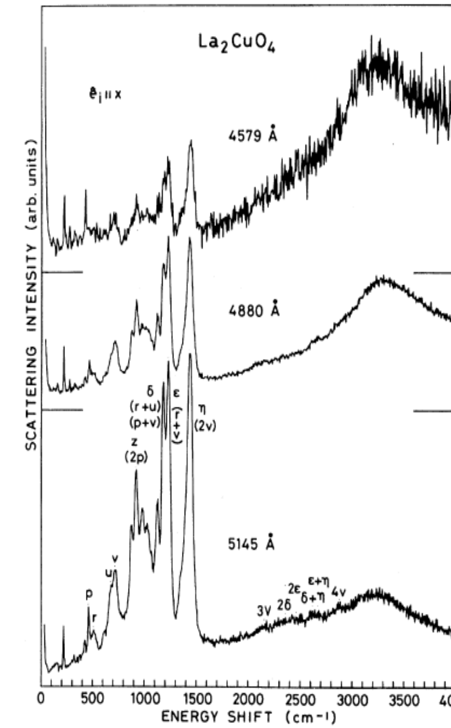
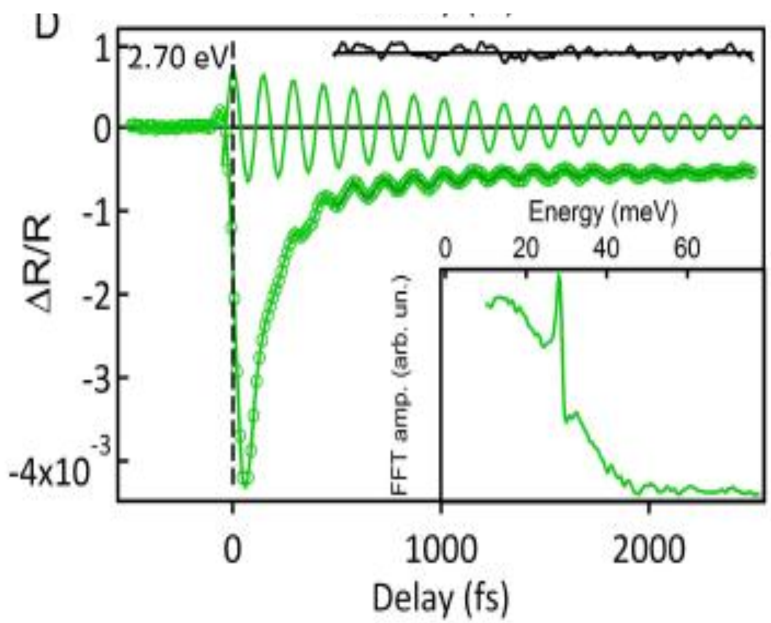
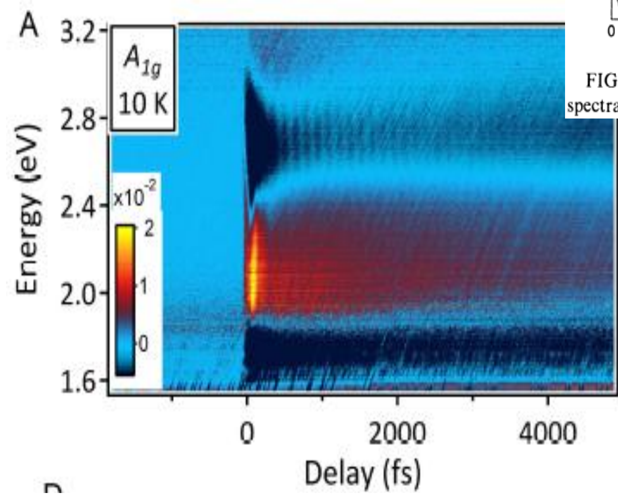


FIG. 10. Incident wavelength dependence of the Raman spectra in La$_2$CuO$_4$ at 30 K.





Real time Raman vs Frequency Domain

Example: Two magnon oscillations in an AF system

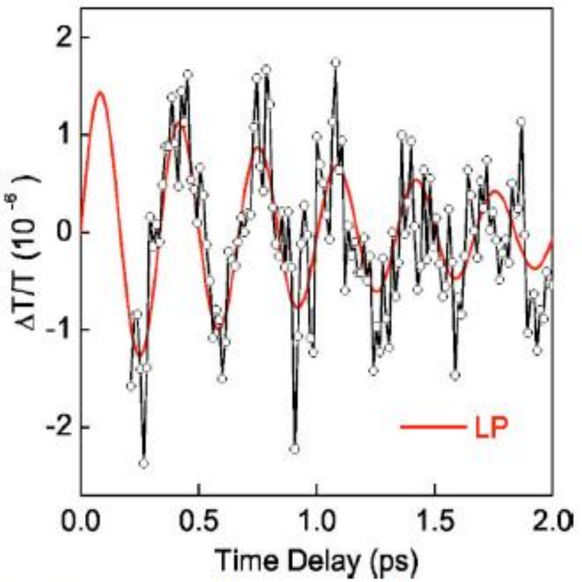
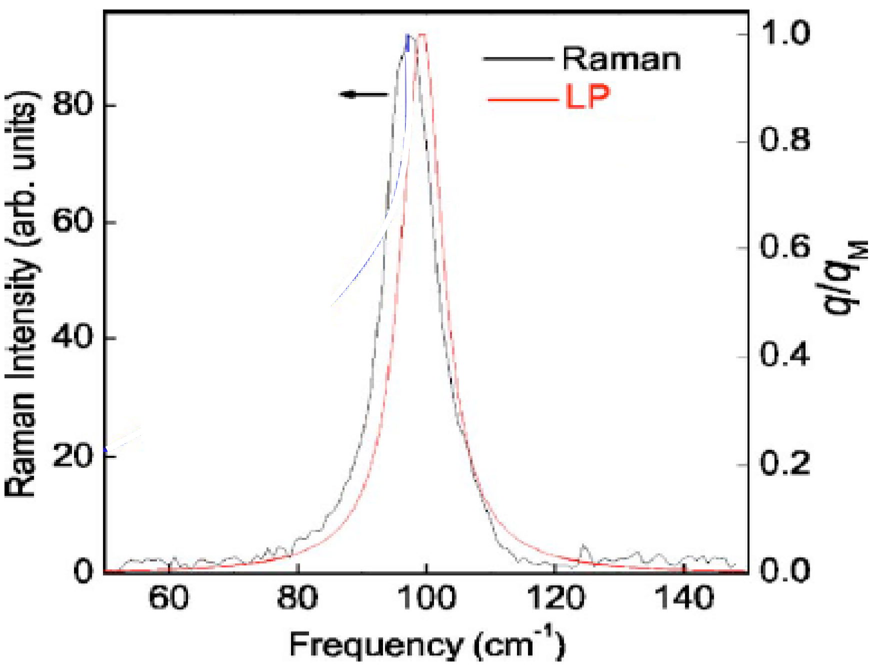


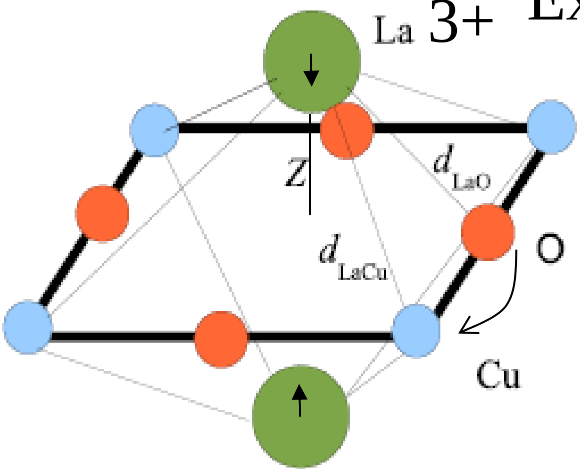
FIG. 9. (Color online) Pump-probe data showing two-magnon oscillations in MnF_2 at 4 K after removal of the phonon oscillation with the linear prediction method. The red line is the linear prediction (LP) model fit.

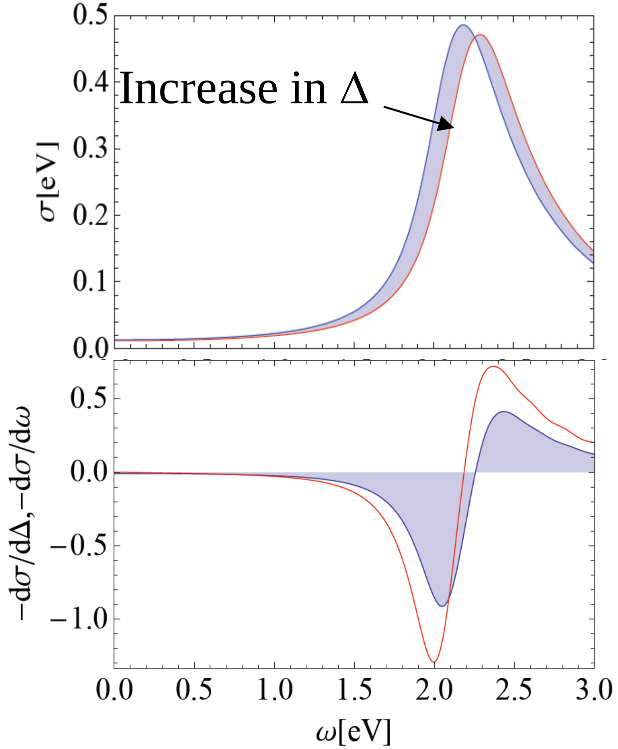


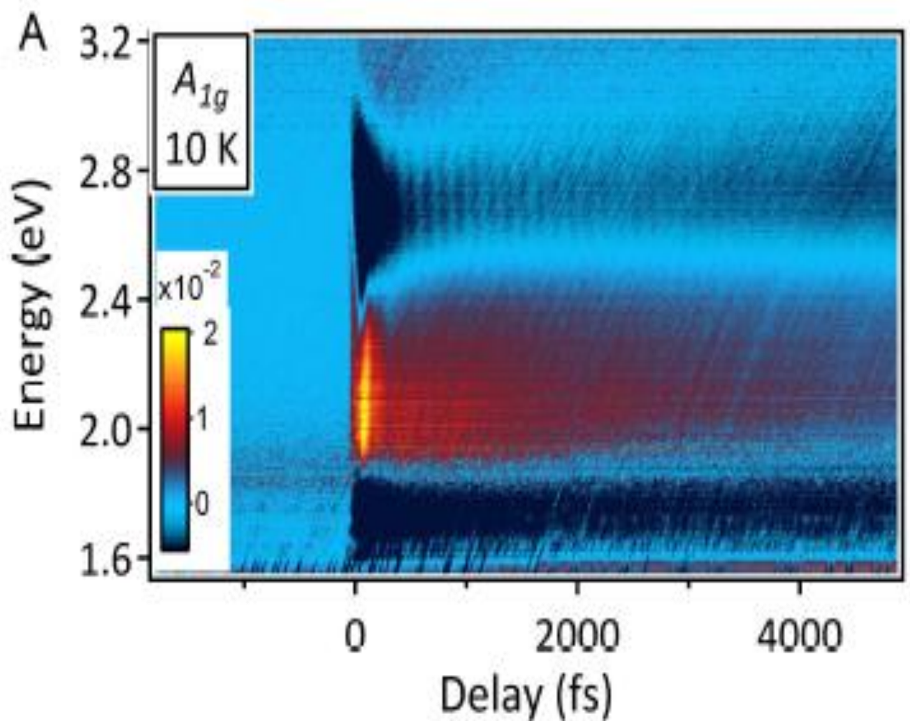
Zhao,Bragas, Merlin, Lockwood PRB 2006

Raman profile as a fingerprint of excitations

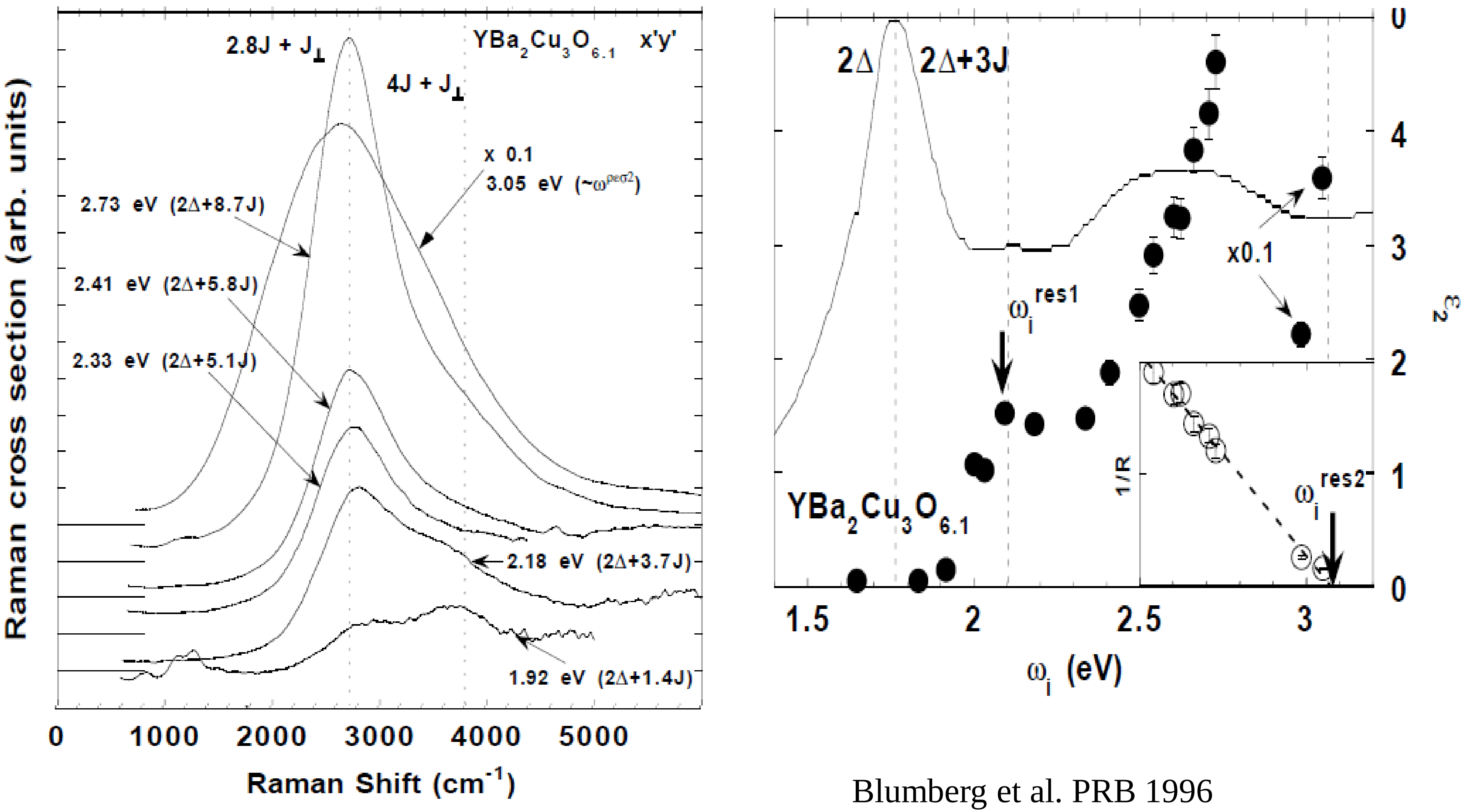
Example: The A1g La Phonon





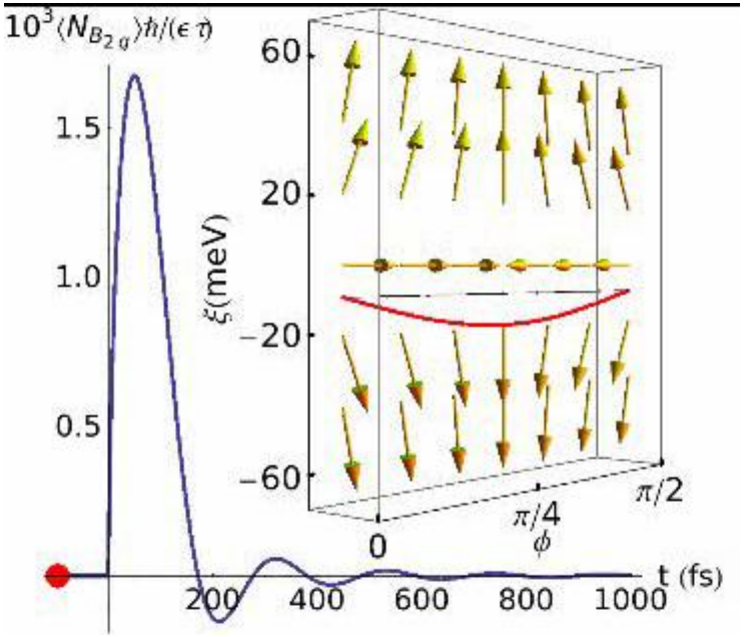


Resonant Effects in Electronic Raman Scattering

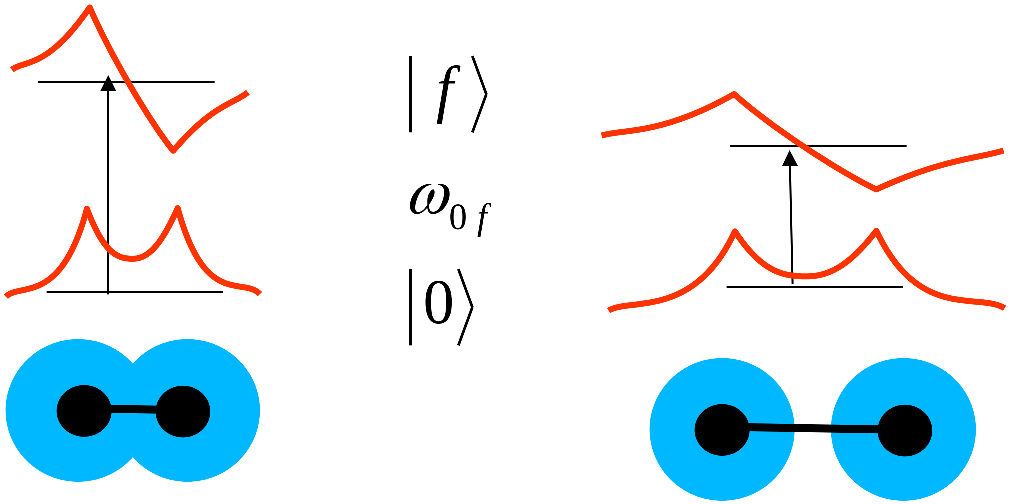


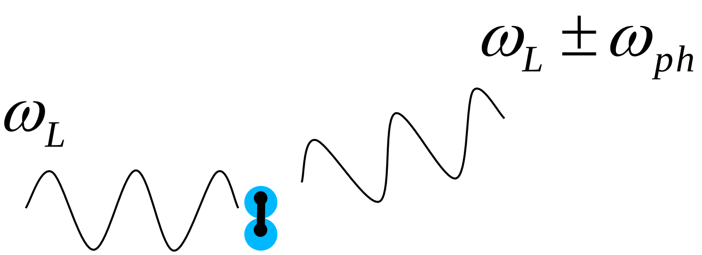
Blumberg et al. PRB 1996

NMR in Charge Space



Raman Scattering

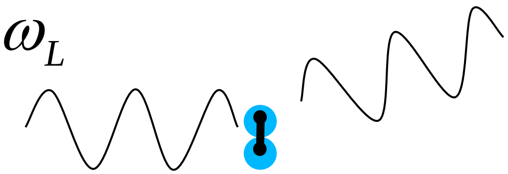




Raman Scattering

$$\xi(t) = \xi e^{-i\omega_{ph}t}$$

$$\sqrt{M_i}\mathbf{u}_i = \mathbf{e}_i \xi e^{-i\omega_{ph}t} + \mathbf{e}_i^* \xi^* e^{i\omega_{ph}t}$$



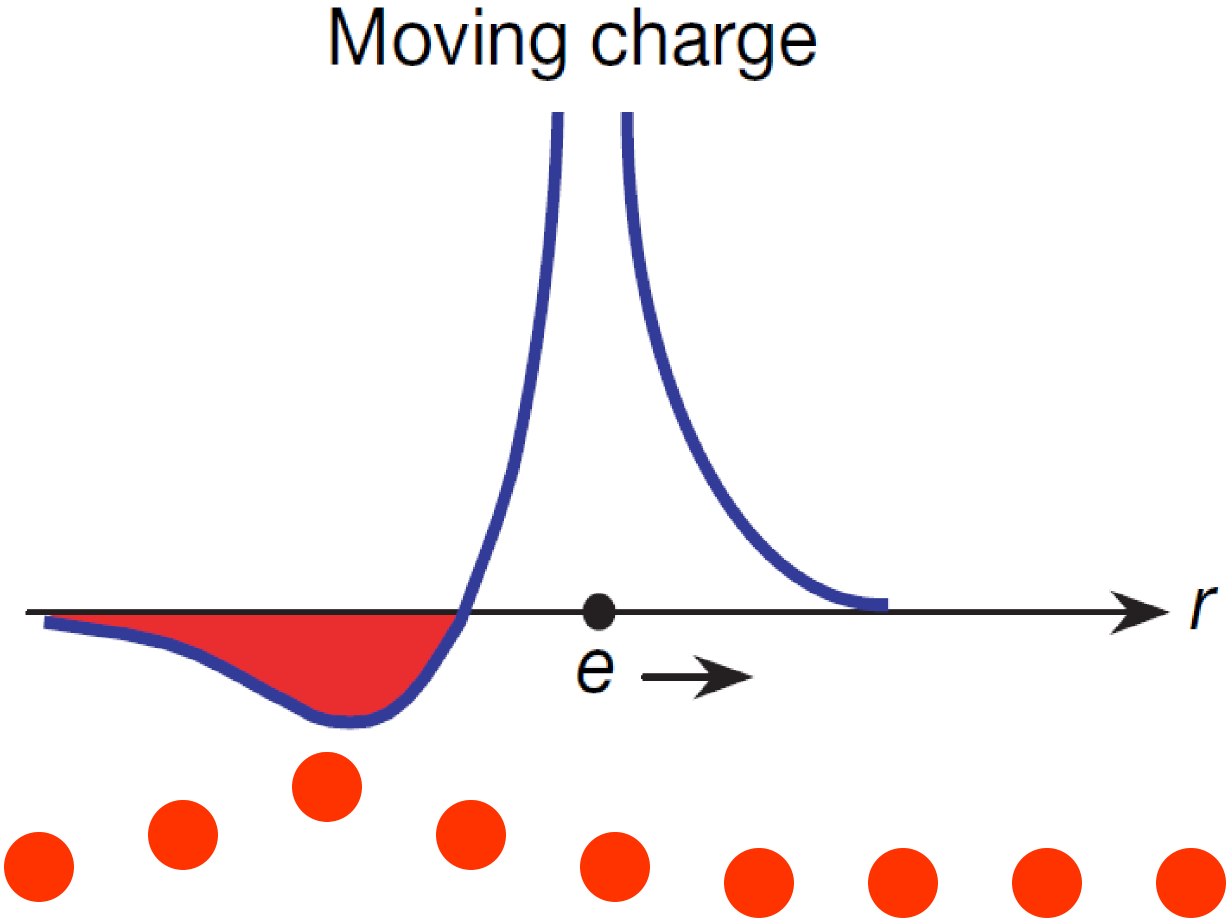
$$\boldsymbol{\alpha} = \frac{e^2}{m} \frac{\mathbf{F}}{\omega_{0f}^2 - \omega^2 - i\omega\gamma}$$

$$\boldsymbol{\alpha} = \boldsymbol{\alpha}_0 + \frac{\partial \boldsymbol{\alpha}}{\partial \xi} \xi e^{-i\omega_{ph}t} + \frac{\partial \boldsymbol{\alpha}}{\partial \xi^*} \xi^* e^{i\omega_{ph}t}$$

$$\mathbf{p} = \boldsymbol{\alpha}.\mathbf{E} \qquad \mathbf{E} = \mathbf{E}_0 e^{-i\omega_L t}$$

$$\mathbf{p} = \left(\boldsymbol{\alpha}_0 e^{-i\omega_L t} + \frac{\partial \boldsymbol{\alpha}}{\partial \xi} \xi e^{-i(\omega_L + \omega_{ph})t} + \frac{\partial \boldsymbol{\alpha}}{\partial \xi^*} \xi^* e^{-i(\omega_L - \omega_{ph})t} \right) \mathbf{E}_0$$

Conventional Superconductors



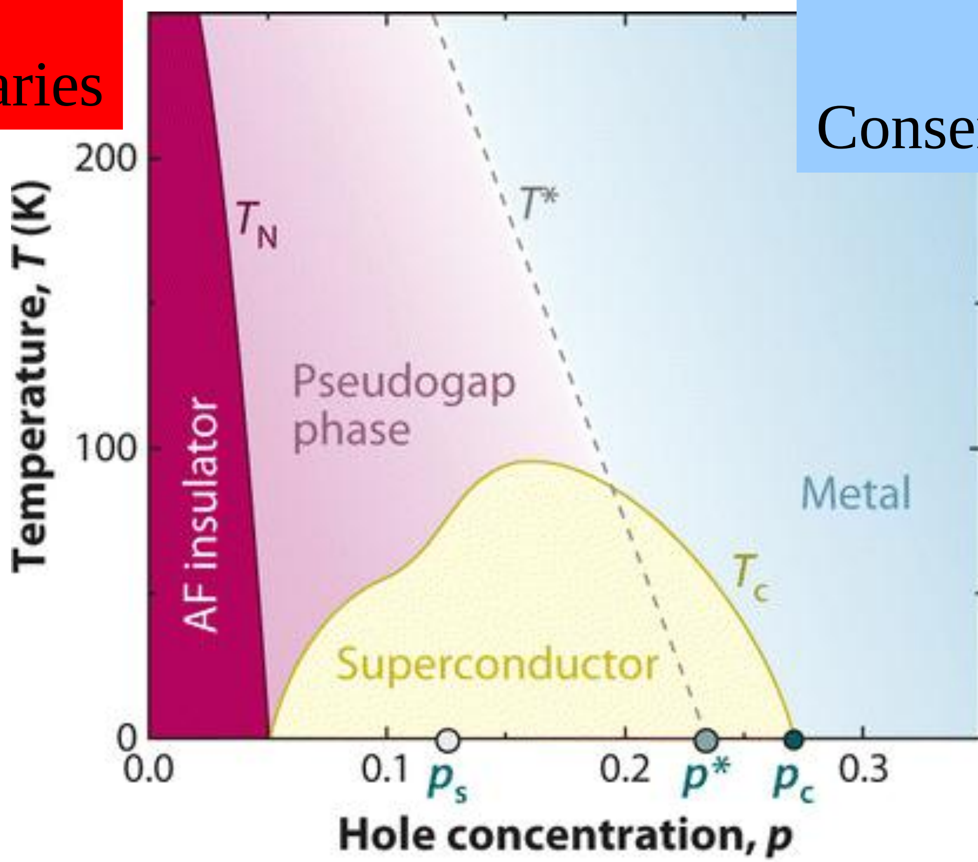
High-Tc Cuprates

Correlated Insulator

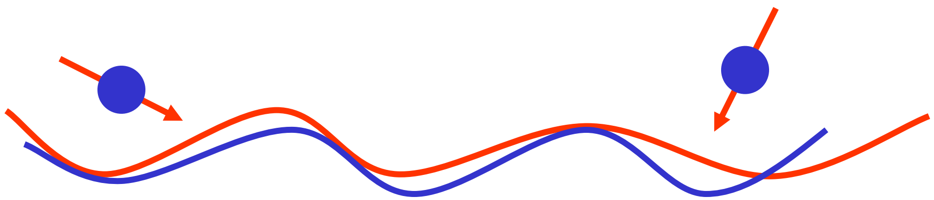
Fermi Liquid

Left Revolutionaries

Right Conservatives/Reformists



Conservative/Reformist View



$$V_{\text{tot}}(\mathbf{r},t) = V_{\text{dir}}(\mathbf{r},t) + V_{\text{ind}}(\mathbf{r},t)$$

$$V_{\text{ind}}(\mathbf{r},t) = -e\,e'\,g_n^2\,\chi_n(\boldsymbol{r},t) - \boldsymbol{s}\bullet\boldsymbol{s}'\,g_m^2\,\chi_m(\boldsymbol{r},t)$$

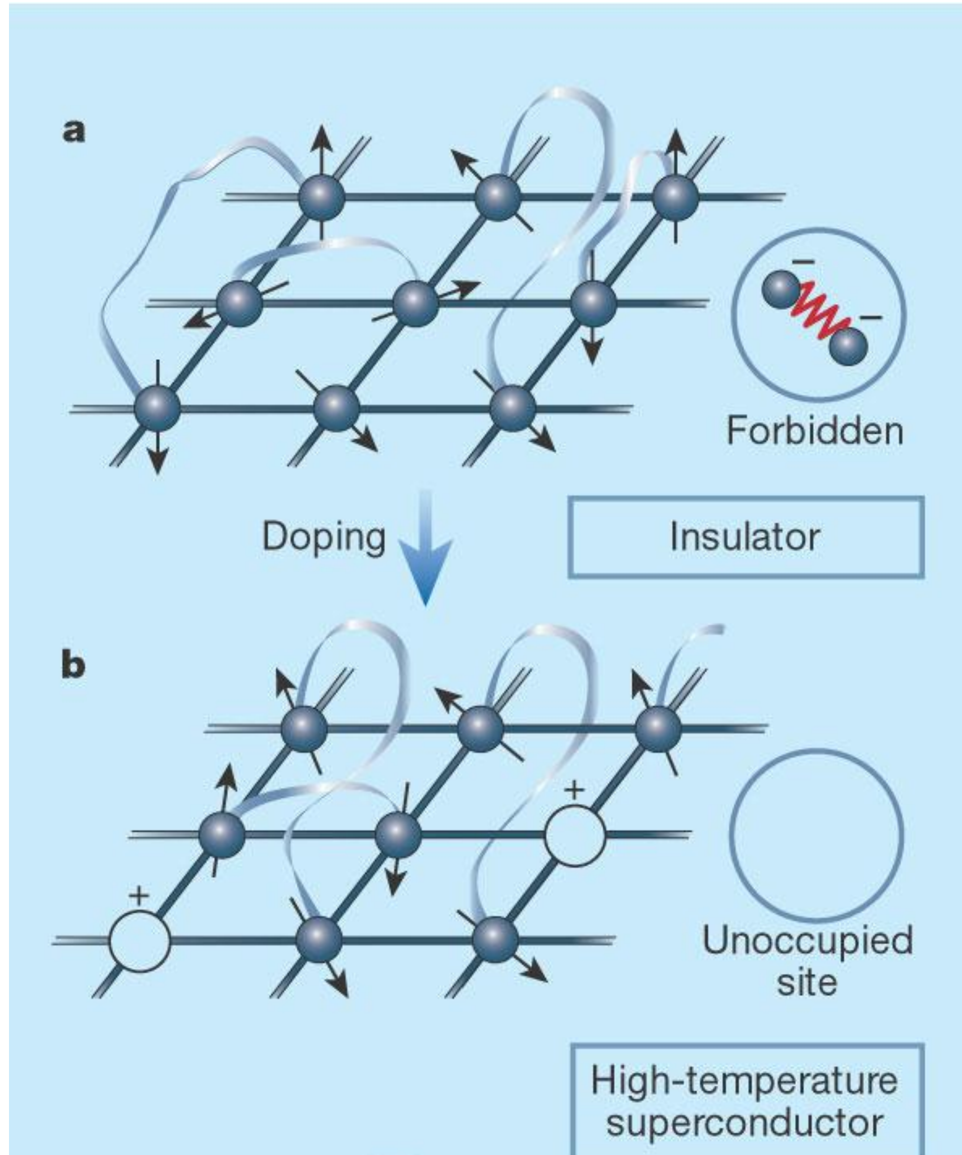
^{3}He: Leggett, RMP 1975

Superconductors: Monthoux, Pines, Lonzarich, Nature 2007

“Left Revolutionary”

PW Anderson

RVB
Scales U and J
Important
No retardation



Raman in Solids

$$\mathbf{P} = n\mathbf{p} \qquad \boldsymbol{\chi} = n\boldsymbol{\alpha} \qquad \mathbf{P} = \boldsymbol{\chi}.\mathbf{E} \qquad \boldsymbol{\varepsilon} = 1 - 4\pi\boldsymbol{\chi} \qquad \boldsymbol{\chi} = \frac{\boldsymbol{\sigma}}{i\omega}$$

$$\boldsymbol{\chi} = \boldsymbol{\chi}_0 + \frac{\partial \boldsymbol{\chi}}{\partial \xi}\xi e^{-i\omega_{ph}t} + \frac{\partial \boldsymbol{\chi}}{\partial \xi^*}\xi^* e^{i\omega_{ph}t}$$

$$\omega_R = \omega_L - \omega_s$$

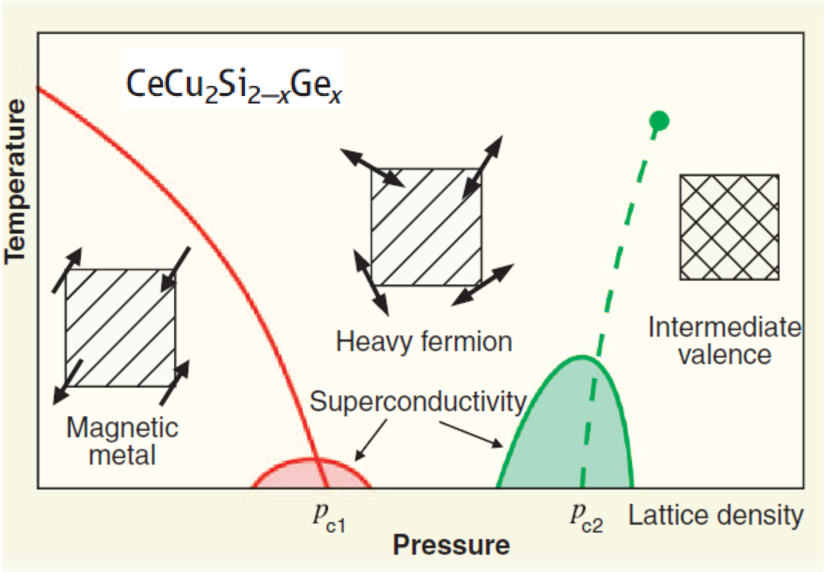
$$\frac{d\sigma}{d\Omega d\omega} = \frac{\omega_s^4 V^2}{(4\pi)^2 c^4} \underbrace{\sum_\nu |\langle 0|\hat{e}_s.\hat{\rho}.\hat{e}_l|\nu\rangle|^2 \delta(\omega_R - \omega_\nu)}_{\frac{1}{\pi}\mathrm{Im}\,\Pi(\omega_R)}$$

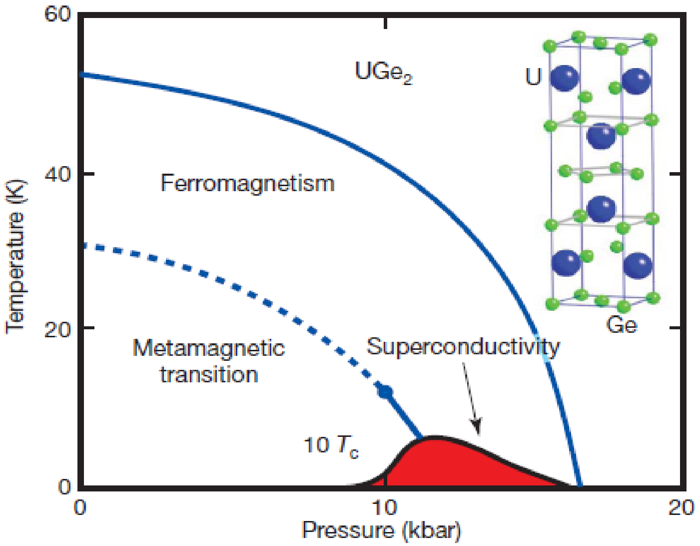
$$\hat{\boldsymbol{\rho}} = \frac{\partial \boldsymbol{\chi}}{\partial \xi}(\omega_L)\xi \qquad \hat{\rho}_{\mu\nu} = \sum_{yX} \frac{\partial \chi_{\mu\nu}}{\partial N_{yX}}(\omega_L)\hat{N}_{yX}$$

$$\Pi(\omega) = i\int_{-\infty}^{t} dt' e^{i\omega t'} \left\langle \left[\hat{e}_L.\hat{\boldsymbol{\rho}}(t).\hat{e}_s, \hat{e}_L.\hat{\boldsymbol{\rho}}(t').\hat{e}_s\right]\right\rangle$$

Cardona, Light Scattering in Solids II

Heavy Fermions



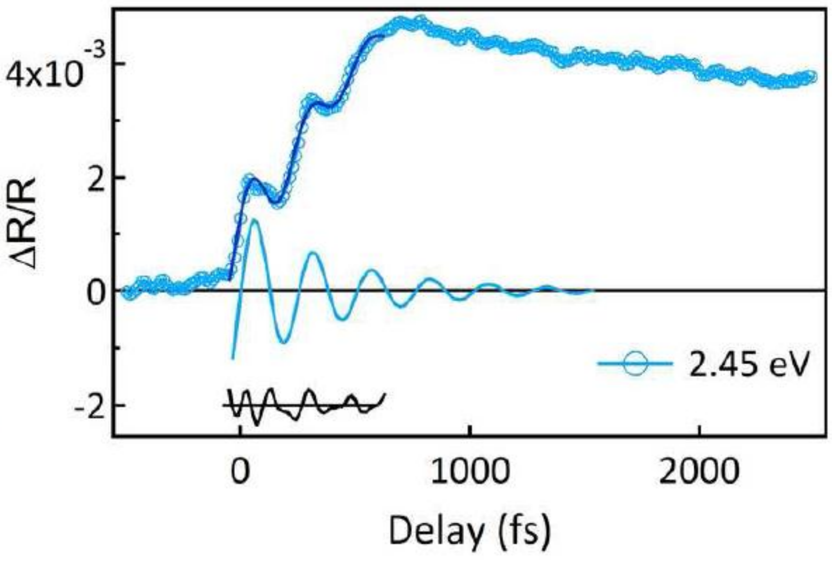


Coherent Excitation of charge fluctuations by Impulsive Stimulated Raman Scattering

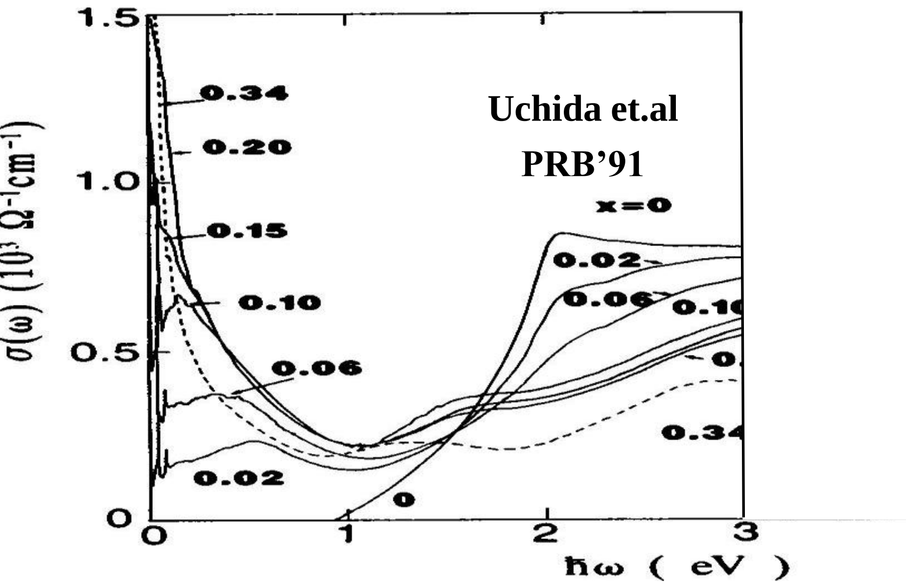
$$H_R(t) = -\frac{1}{2}\sum_k \mathbf{E}(t).\chi_{el}^R.\mathbf{E}(t) f_k^X (n_{k\uparrow} + n_{-k\downarrow})$$

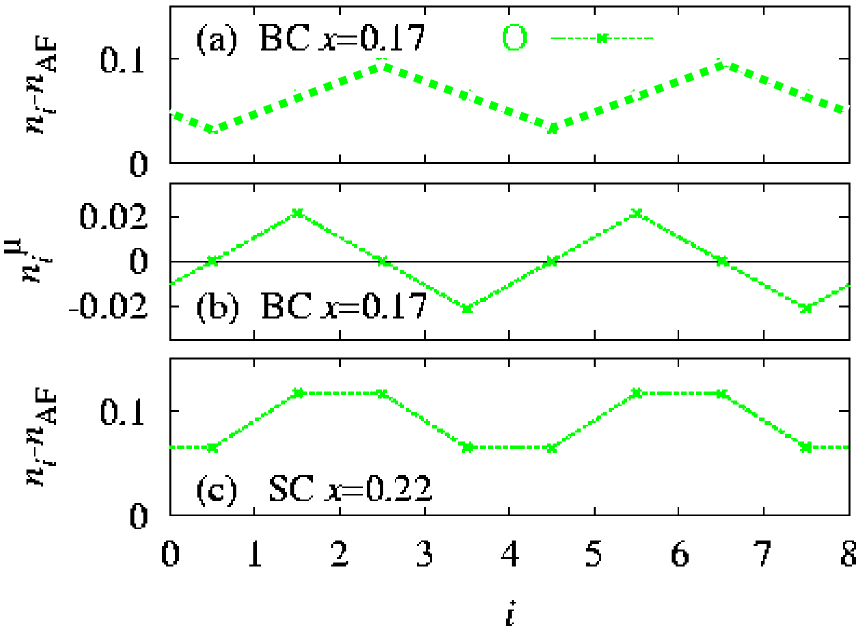
$$H_R(t) = \sum_k v_k^X(t)(n_{k\uparrow} + n_{-k\downarrow})$$

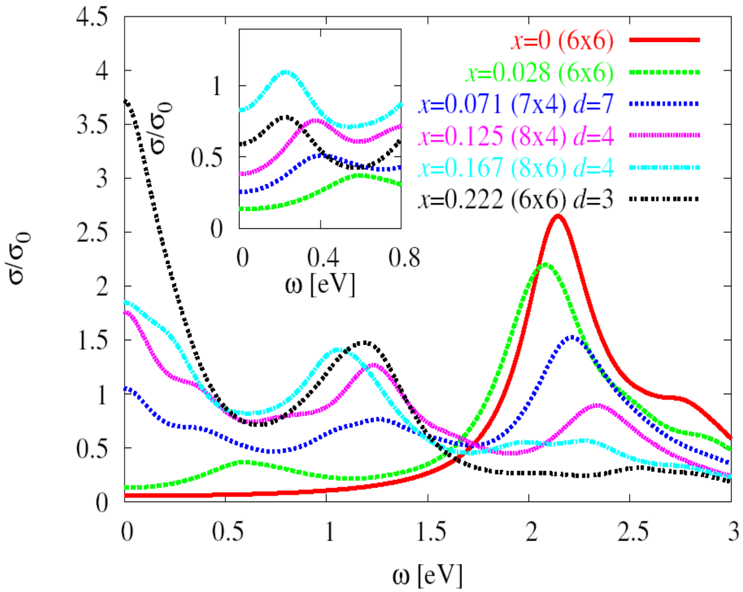
$$v_k^X(t) = -\frac{1}{2}\mathbf{E}(t).\chi_{el}^R.\mathbf{E}(t) f_k^X$$

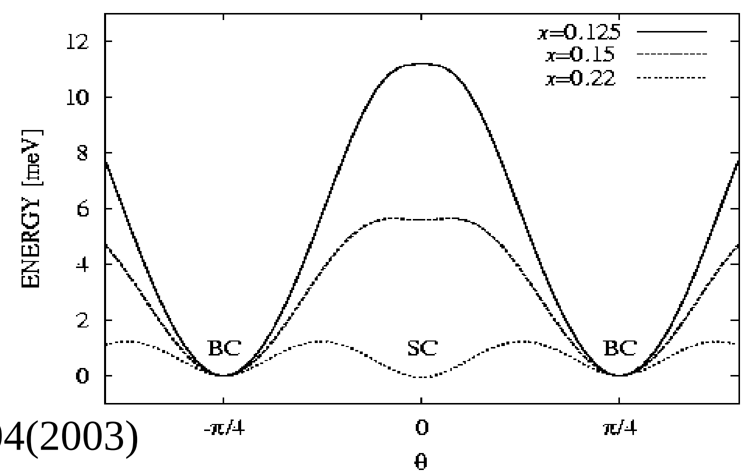


Optical Conductivity and Fluctuations







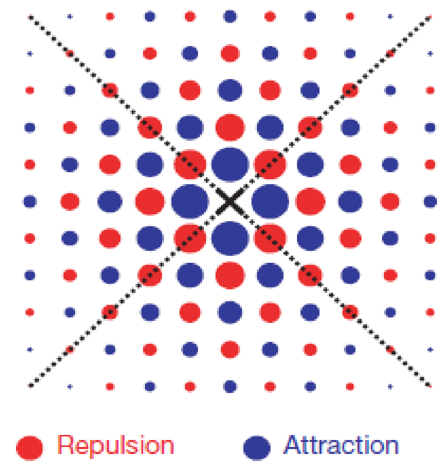


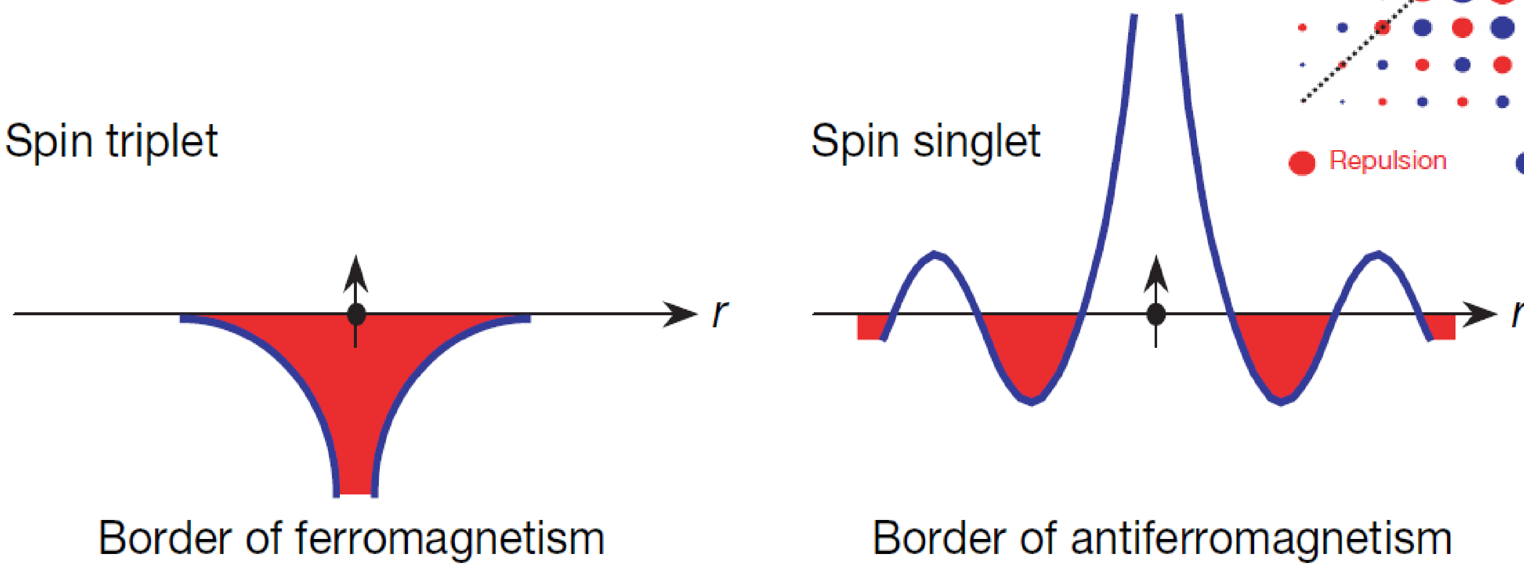
J.L & G.S., PRL **90**, 066404(2003)

Unconventional Superconductors

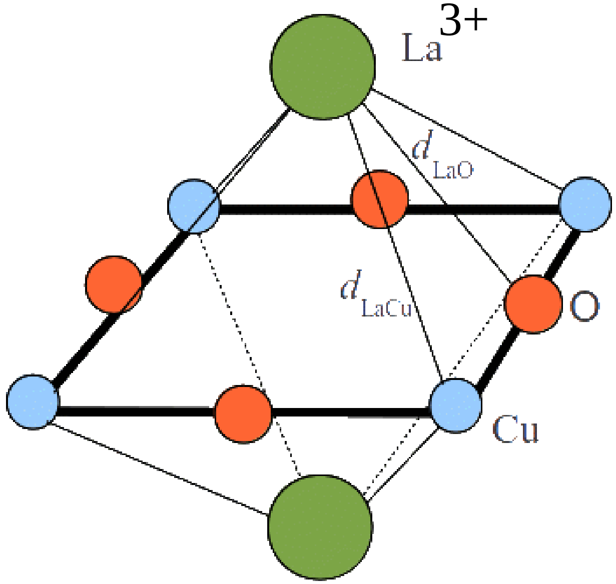
$$V_{\mathrm{ind}}(\mathbf{r},t) = -\, e\, e'\; g_n{}^2\, \chi_n(\boldsymbol{r},\, t) - \boldsymbol{s}\bullet\boldsymbol{s}'\; g_m{}^2\, \chi_m(\boldsymbol{r},t)$$

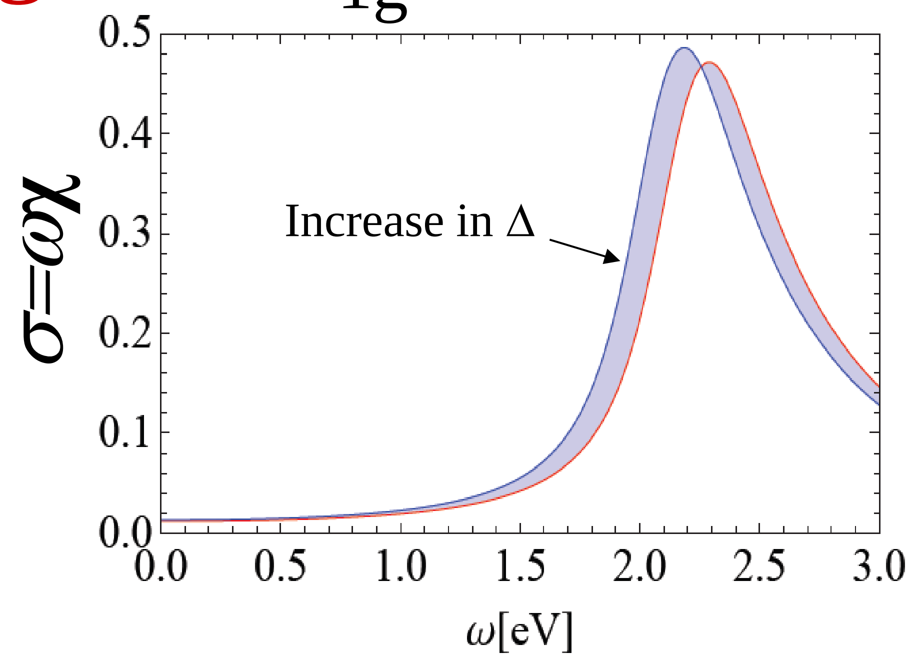
Close to instabilities the susceptibility is large at small energies (long times)





Raman Scattering: the A_{1g} La Phonon

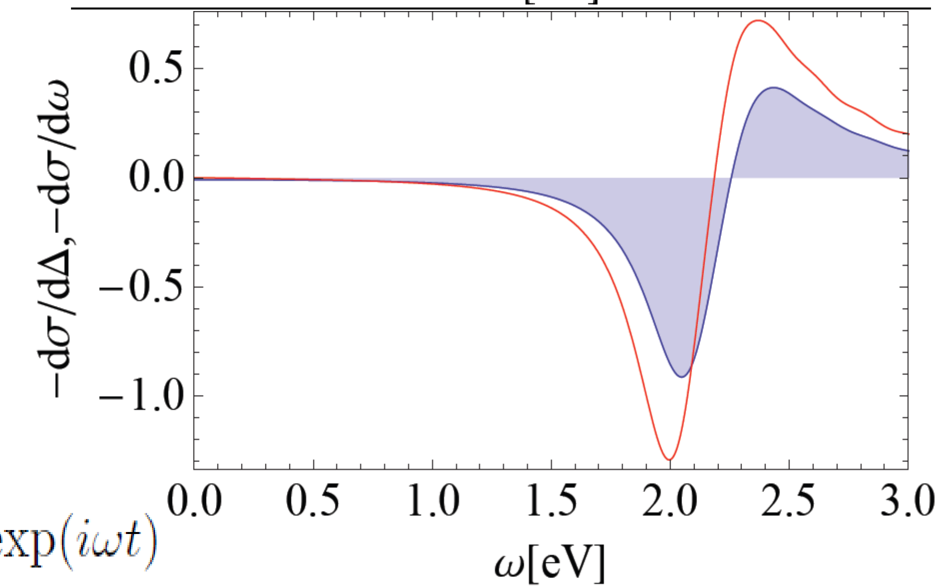




$$\Delta = \Delta_0 + Ze^2\left(\frac{1}{d_{LaO}} - \frac{1}{d_{LaCu}}\right)$$

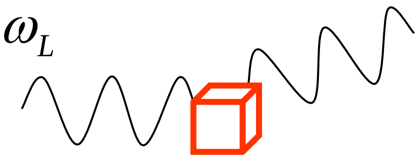
el-ph

$$\frac{d\chi}{d\xi} = \frac{d\chi}{d\Delta}\frac{d\Delta}{dz}\frac{dz}{d\xi} \qquad \frac{d\chi}{d\Delta} \approx -\frac{d\chi}{d\omega}$$



$$\sqrt{M_{La}}\mathbf{u}_{La} = \mathbf{e}_{La}\xi\exp(-i\omega t) + \mathbf{e}^*_{La}\xi^*\exp(i\omega t)$$

Raman Scattering



$$H = H_{ph} - \frac{1}{2}\mathbf{E}(t).\frac{\partial\chi(\omega_L)}{\partial\xi}.\mathbf{E}(t)\hat{\xi} = H_{ph} - F(t)\hat{\xi}$$

$$\frac{d\sigma}{d\Omega d\omega} = \frac{\omega_s^4 V^2}{(4\pi)^2 c^4}\underbrace{\sum_\nu |\langle 0|\hat{e}_s.\hat{\rho}.\hat{e}_l|\nu\rangle|^2\delta(\omega_R - \omega_\nu)}_{\frac{1}{\pi}\mathrm{Im}\,\Pi(\omega_R)}$$

$$\hat{\boldsymbol{\rho}} = \frac{\partial\boldsymbol{\chi}}{\partial\xi}(\omega_L)\xi$$

$$\Pi(\omega) = i\int_{-\infty}^{t} dt' e^{i\omega t'}\langle[\hat{e}_L.\hat{\boldsymbol{\rho}}(t).\hat{e}_s, \hat{e}_L.\hat{\boldsymbol{\rho}}(t').\hat{e}_s]\rangle$$

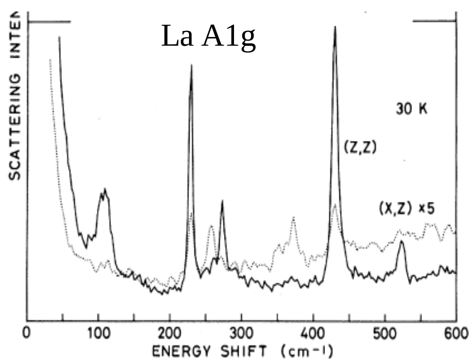


FIG. 9. Raman spectra of $(La_{1-x}Sr_x)_2CuO_4$ with $x = 0.035$ at 30 and 337 K in the (z,z) (solid curves) and (x,z) (dotted curves) polarization configurations.

Sugai PRB ‘89

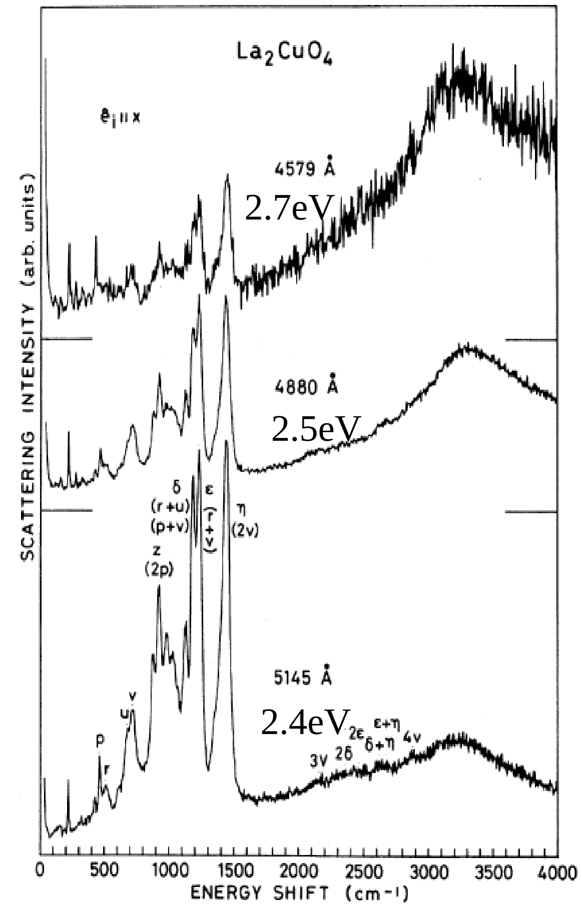


FIG. 10. Incident wavelength dependence of the Raman spectra in La_2CuO_4 at 30 K.