Energy versus time in x-ray scattering

Peter Abbamonte
University of Illinois

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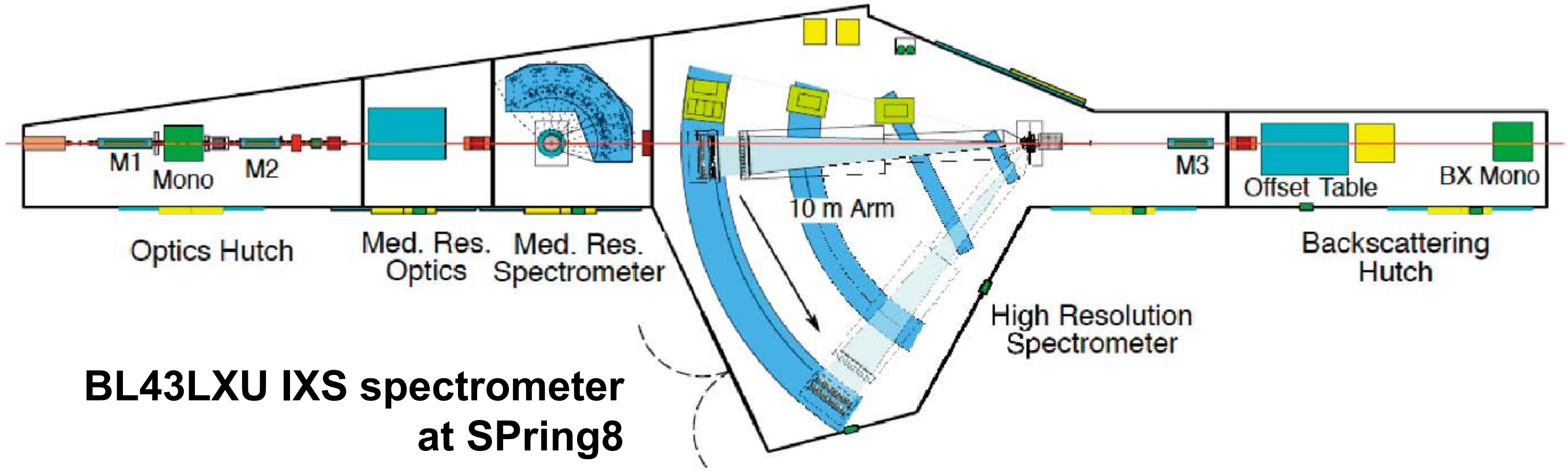
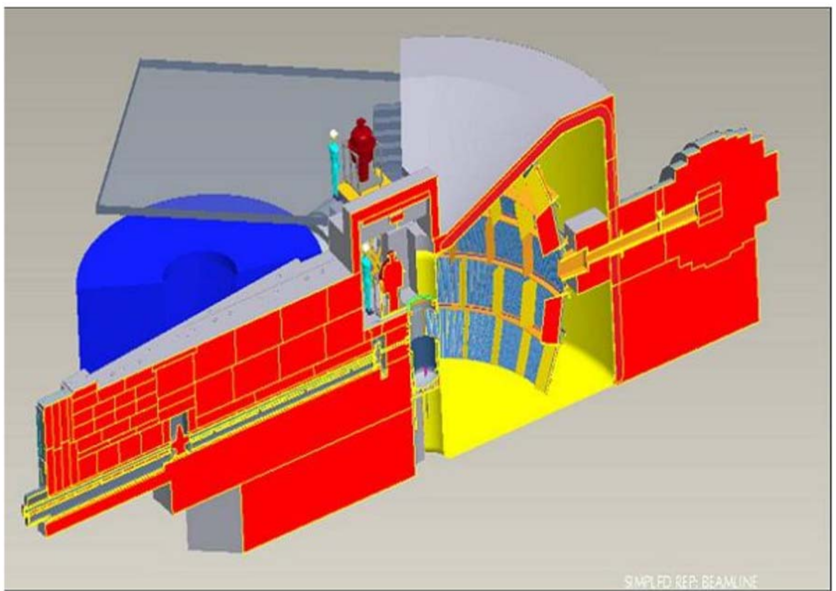
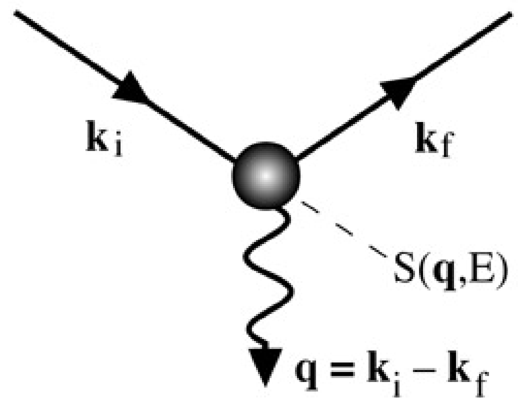
Momentum- and energy-resolved scattering



BL43LXU IXS spectrometer at SPring8



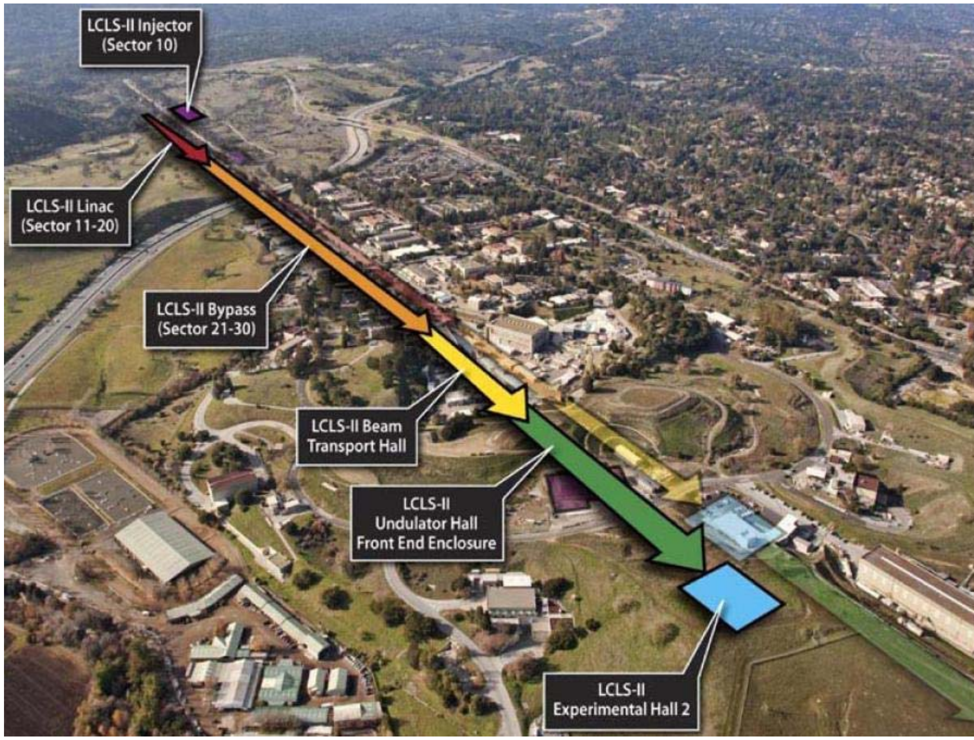
SEQUOIA spectrometer at SNS

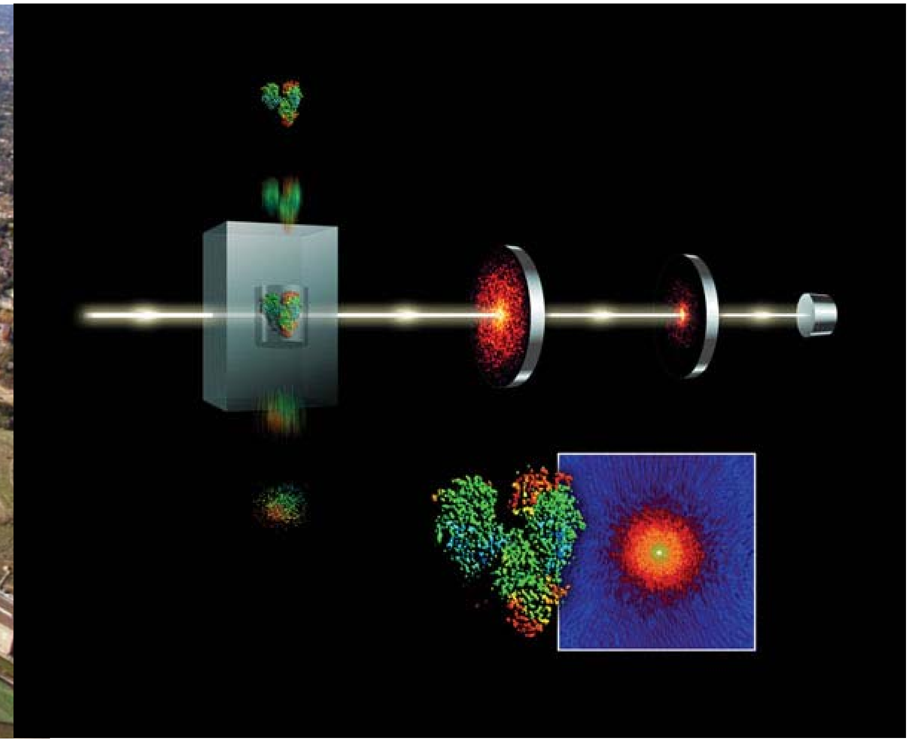


Measures a correlation function, S(q,ω)

(See talk yesterday by Toby Perring)

A new approach: Free Electron Lasers





Can—for the first time—study ultrafast dynamics with a momentum-resolved probe

Questions for today:

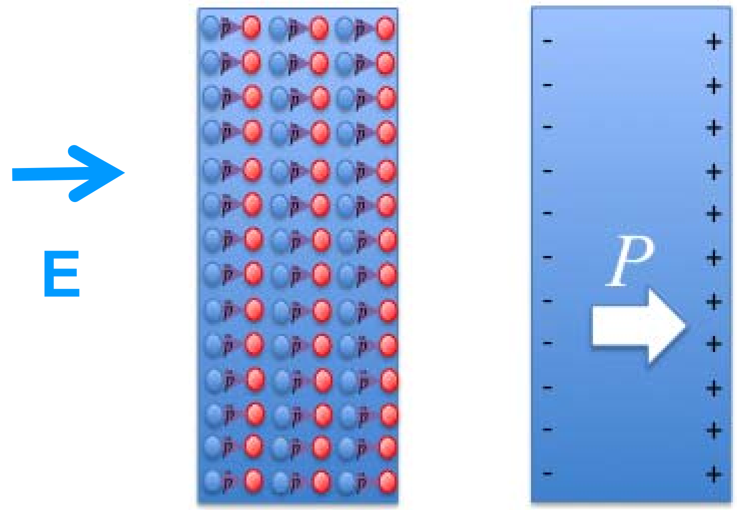
- **How is this different from inelastic scattering techniques, which are also said to measure dynamics? That is, how are time and frequency related?**
- **Where does scattering come from, and how does it measure dynamics anyway?**

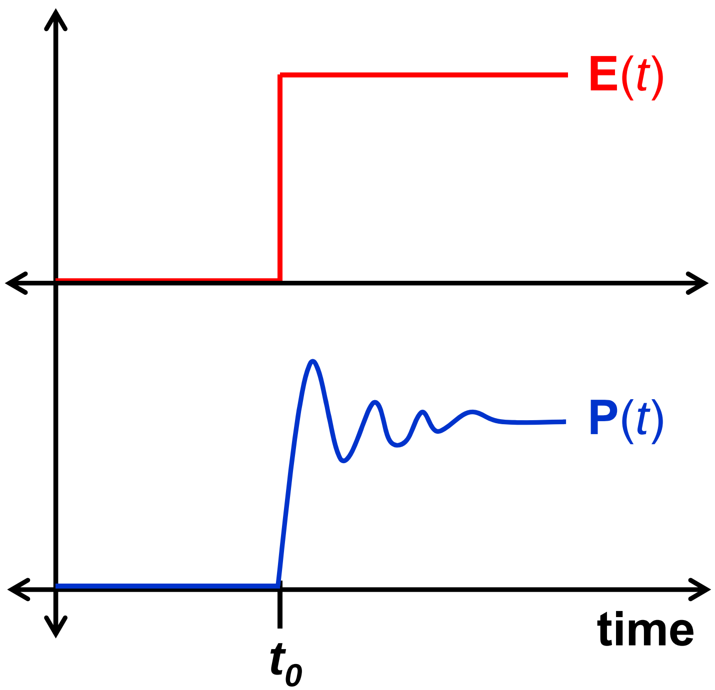
Example of ω vs. t : the dielectric function, $\varepsilon(\omega)$

$$\varepsilon(\omega) = 1 + 4\pi\chi_e(\omega)$$

$$\chi_e(\omega) = \frac{\varepsilon(\omega) - 1}{4\pi}$$

$$\mathbf{P}(\omega) = \chi_e(\omega)\mathbf{E}(\omega)$$





Time relationship is nonlocal:

$$\chi_e(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \chi_e(\omega) e^{-i\omega t}$$

$$\mathbf{P}(t) = \int_{-\infty}^{\infty} dt' \chi_e(t - t')\mathbf{E}(t')$$

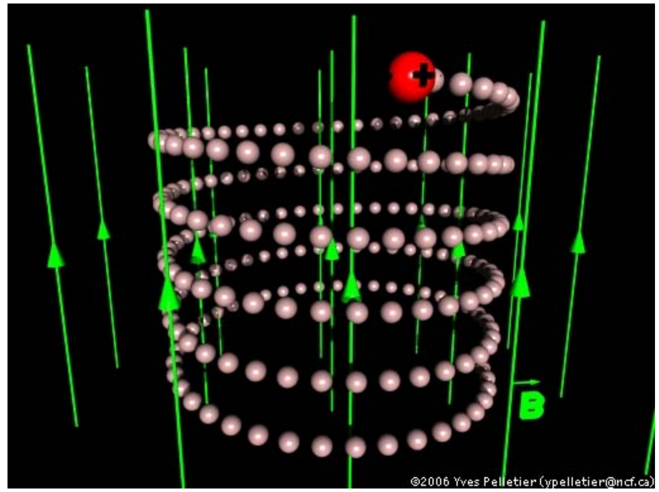
χ_e is a Green's function

ω dependence $\leftrightarrow$ retardation

Electron in an EM field (classical)

Can define the fields in terms of potentials:

$$\mathbf{B} = \nabla \times \mathbf{A} \qquad\qquad \mathbf{E} = -\nabla \varphi - \frac{1}{c}\frac{\partial \mathbf{A}}{\partial t}$$



Classical motion described by the Lagrangian

$$L = K - V = \frac{1}{2}m\dot{\mathbf{x}}^2 + e\varphi - \frac{e}{c}\dot{\mathbf{x}} \cdot \mathbf{A} \; {}^{*}$$

The canonical momentum is

$$\mathbf{p}^c = \frac{\partial L}{\partial \dot{\mathbf{x}}} = m\dot{\mathbf{x}} - \frac{e}{c}\mathbf{A}$$

This allows one to define the classical Hamiltonian

$$H = \sum_i p_i^c \dot{r}_i - L = \frac{1}{2m}\left(\mathbf{p}^c + \frac{e}{c}\mathbf{A}\right)^2 - e\varphi$$

Hamilton's equations give the equations of motion:

$$\dot{x}_i = \frac{\partial H}{\partial p_i^c} \qquad\qquad \dot{p}_i = -\frac{\partial H}{\partial x_i}$$

Result is the Lorentz force law:

$$m\ddot{\mathbf{x}} = -e\left(\mathbf{E} + \frac{1}{c}\dot{\mathbf{x}} \times \mathbf{B}\right)$$

* **Gaussian units**

Electron in an EM field (quantum)

The Hamiltonian is now an operator. Photons are massless so we have to use second quantization:

$$\hat{H} = \hat{H}_{EM} + \hat{H}_{\text{electron}} + \hat{H}_{\text{interaction}}$$

$$\hat{H}_{EM} = \int d\mathbf{x}\left(\frac{\hat{E}^2}{2} + \frac{\hat{B}^2}{2}\right) \qquad \hat{H}_{electron} = \int d\mathbf{x}^3\, \hat{\psi}^\dagger(\mathbf{x},t)\left[\frac{\mathbf{p}^2}{2m} + V(\mathbf{x})\right]\hat{\psi}(\mathbf{x},t)$$

note:
$$\mathbf{p} = m\dot{\mathbf{x}} - \frac{e}{c}\mathbf{A}$$

Where $\hat{\psi}(\mathbf{x},t)$ annihilates an electron at position *x* and time *t*.

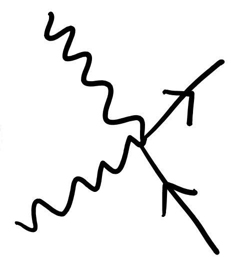
The vector potential is an operator that creates or annihilates photons:

$$\hat{\mathbf{A}}(\mathbf{x},t) = \sum_{k,\lambda} c\sqrt{\frac{\hbar}{2\omega_k}}\left[a_{k,\lambda}\,\epsilon_\lambda\, e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} + a^\dagger_{k,\lambda}\,\epsilon^*_\lambda\, e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega t)}\right]$$

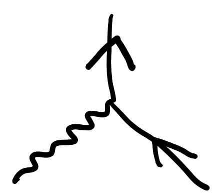
Multiplying everything out gives fundamental interactions between electrons and photons:

$$H = H_0 + H_1 + H_2$$

$$H_2^I(t) = -\frac{e^2}{2mc^2}\int d\mathbf{x}^3\, \hat{\psi}^\dagger(\mathbf{x},t)\ \hat{\mathbf{A}}^2(\mathbf{x},t)\ \hat{\psi}(\mathbf{x},t) = -\frac{e^2}{mc^2}\int d\mathbf{x}^3\ \hat{\mathbf{A}}^2(\mathbf{x},t)\ \hat{n}(\mathbf{x},t)$$



$$H_1^I(t) = -\frac{e}{mc}\int d\mathbf{x}^3\, \hat{\psi}^\dagger(\mathbf{x},t)\left[\mathbf{p}\cdot\hat{\mathbf{A}}(\mathbf{x},t)\right]\hat{\psi}(\mathbf{x},t)$$



Scattering

Scattering takes place when these interactions evolve a photon from an initial state to a final state, with a corresponding change in the electronic subsystem:

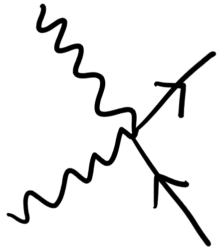
$$|i\rangle = a^{\dagger}_{k_i,\lambda_i}|m\rangle \longrightarrow |f\rangle = a^{\dagger}_{k_f,\lambda_f}|n\rangle$$

What does this is the time-evolution operator:

$$U_I(\infty,-\infty) = \exp\left[-i\int_{-\infty}^{\infty} dt\, H^I(t)\; e^{-\eta|t|}\right] \qquad M = \langle i|U_I(\infty,-\infty)|f\rangle$$

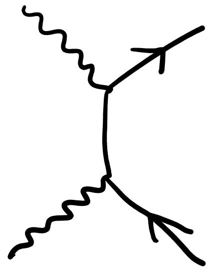
"Nonresonant" x-ray scattering

$$w_{f\leftarrow i} = r_0^2(\epsilon_f^* \cdot \epsilon_i)^2 \sum_{n,m} |<n|\hat{n}(\mathbf{k})|m>|^2\, P_m\, \delta(\omega - \omega_n + \omega_m)$$



"Resonant" inelastic x-ray scattering (RIXS)

$$w_{f\leftarrow i} = \left|\frac{e^2}{mc^2\hbar^2}\sum_m \frac{<f|\mathbf{p}\cdot\mathbf{A}|m><m|\mathbf{p}\cdot\mathbf{A}|0>}{\omega - \omega_m + i\gamma}\right|^2 \delta(\omega - \omega_f + \omega_0)$$

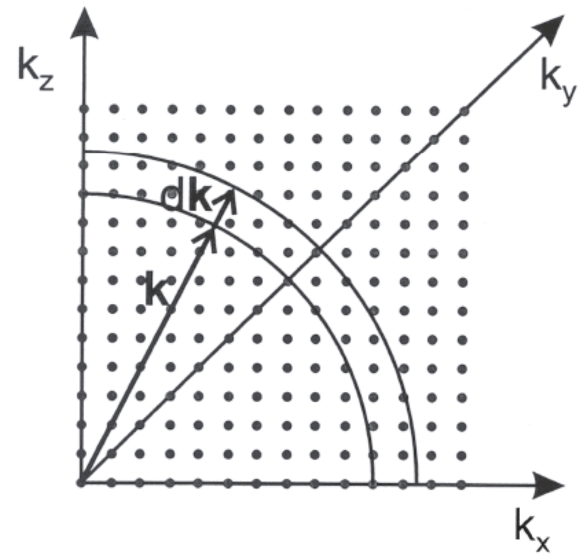


(J. van den Brink, after the coffee break)

Cross section for x-ray scattering

The differential scattering cross section comes from Fermi's golden rule

$$\frac{\partial^2 \sigma}{\partial \Omega \partial E} = \frac{1}{\Phi} w_{f \leftarrow i} \cdot \frac{\partial^2 N}{\partial \Omega \partial E}$$

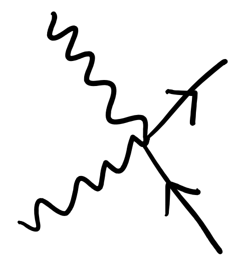


where counting states in a box of volume V provides the density of final states:

$$\frac{\partial^2 N}{\partial \Omega \partial E} = \frac{\omega_f V}{8\pi^3 \hbar c^3}$$

$$\Phi = \frac{c}{V} \quad \text{(for one incident photon)}$$

Cross section for nonresonant x-ray scattering



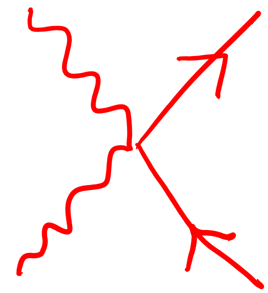
$$\frac{\partial^2 \sigma(\mathbf{q}, \omega)}{\partial \Omega \partial E} = r_0^2 \frac{\omega_f}{\omega_i} \left(\varepsilon_f^* \cdot \varepsilon_i \right)^2 S(\mathbf{q}, \omega)$$

$$\mathbf{k} = \mathbf{k}_f - \mathbf{k}_i \qquad \omega = \omega_f - \omega_i$$

dynamic structure factor – what is it?

$S(\mathbf{q},\omega)$ and the Van Hove function

Cross section:



$$\frac{\partial^2\sigma(\mathbf{q},\omega)}{\partial\Omega\partial E} = r_0^2\,\frac{\omega_2}{\omega_1}\left(\varepsilon_2^*\cdot\varepsilon_1\right)^2 S(\mathbf{q},\omega)$$

Assuming we are in thermodynamic equilibrium, *S* has the form

$$S(\mathbf{q},\omega) = \frac{1}{\hbar}\sum_{m,n} P_m \left|\langle n|\hat{n}(\mathbf{q})|m\rangle\right|^2 \delta(\omega-\omega_n+\omega_m) \qquad P_m = \frac{e^{-\hbar\omega_m/kT}}{Z}$$

This is the so-called "dynamic structure factor."

$S(\mathbf{q},\omega)$ is the Fourier transform of the Van Hove function, *G*(x,*t*), which is the space-time correlation function for the electron density:

$$S(\mathbf{q},\omega) = \int d\mathbf{x}\,dt\,G(\mathbf{x},t)\,e^{-i(\mathbf{q}\cdot\mathbf{x}-\omega t)}$$

where

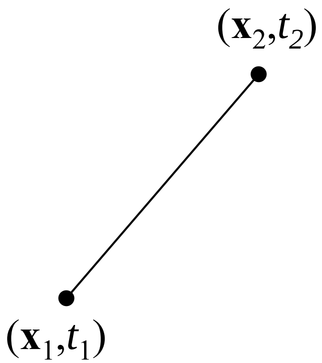
$$G(\mathbf{x},t) = \int d\mathbf{x}'\,dt'\left\langle \hat{n}(\mathbf{x}',t')\,\hat{n}(\mathbf{x}'+\mathbf{x},t'+t)\right\rangle$$

The brackets denote a QM thermal average:

$$\left\langle \hat{O}\right\rangle \equiv \sum_m P_m \langle m|\hat{O}|m\rangle$$

What does this have to do with dynamics?

Fluctuation-Dissipation Theorem

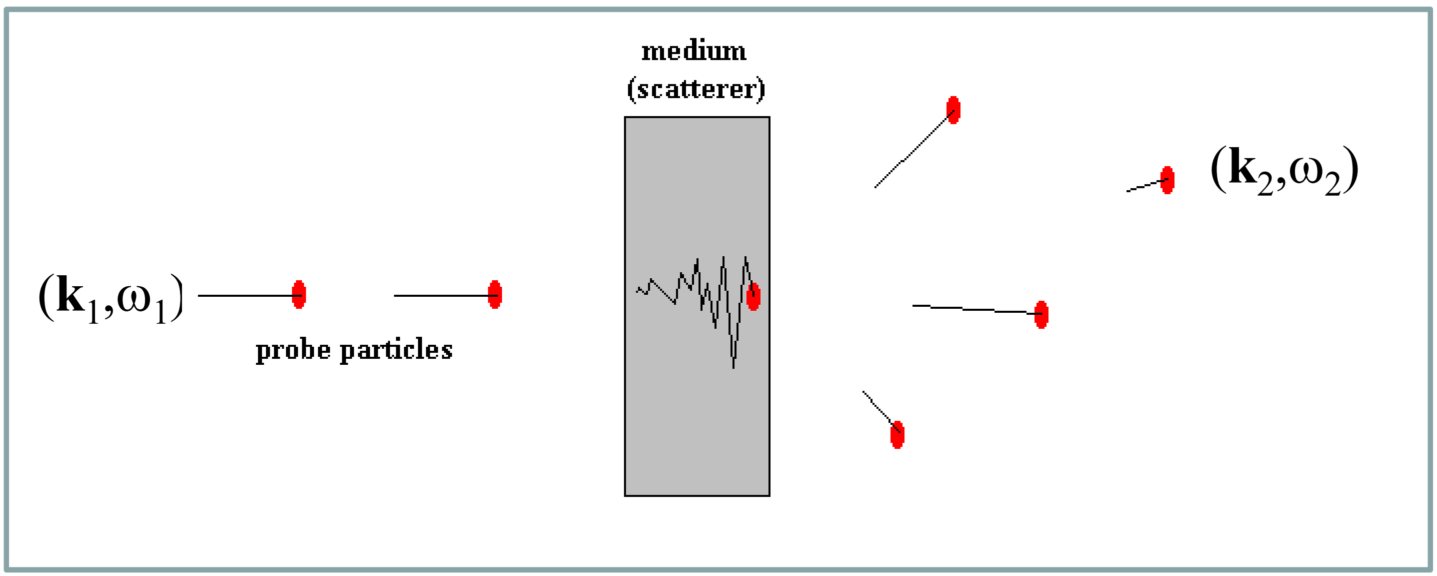


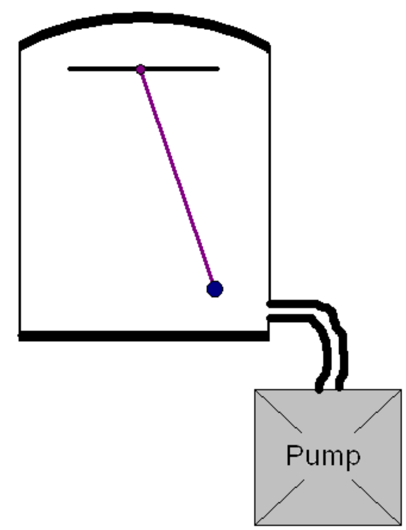
$$S(\mathbf{q},\omega) = -\frac{1}{\pi}\frac{1}{1-e^{-\hbar\omega/kT}}\mathrm{Im}\left[\chi(\mathbf{q},\omega)\right]$$

fluctuation-dissipation theorem

$$\chi(\mathbf{x}_1,\mathbf{x}_2,t_1-t_2) = -\frac{i}{\hbar}\left\langle\left[\hat{\rho}(\mathbf{x}_2,t_2),\hat{\rho}(\mathbf{x}_1,t_1)\right]\right\rangle\theta(t_1-t_2)$$

Retarded density Green's function





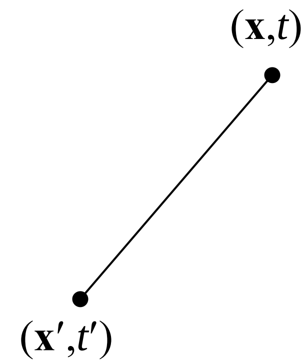
Describes how charge propagates in a system:

- Phonons
- Plasmons
- Excitons
- Electron-hole pairs
- Etc.

Green's functions or "Propagators"

Dynamics is described by a propagator

$$K(\mathbf{x},t;\mathbf{x}',t')$$



Electrons:

$$G(\mathbf{x},t;\mathbf{x}',t') = i/\hbar \left\langle 0 \right| \left\{ \hat{\psi}(\mathbf{x},t), \hat{\psi}^{\dagger}(\mathbf{x}',t') \right\} \left| 0 \right\rangle \theta(t-t')$$

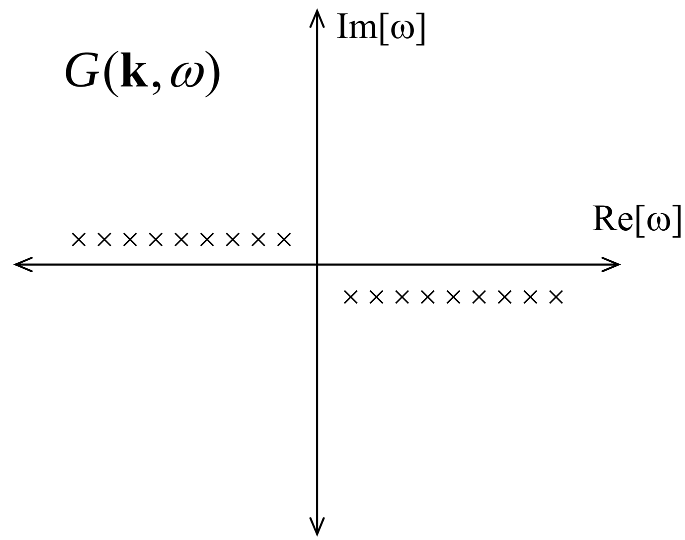
Density:

$$\chi(\mathbf{x},t;\mathbf{x}',t') = i/\hbar \left\langle 0 \right| \left[\hat{\rho}(\mathbf{x},t), \hat{\rho}(\mathbf{x}',t') \right] \left| 0 \right\rangle \theta(t-t')$$

Some translational symmetry:
Frequency / momentum representation is more illuminating.

- **Best way—in a many-body system—to define what is a "particle"**

$$G(\mathbf{k},\omega)$$



View propagator in real time?

Crazy idea: Can we Fourier Transform IXS data and make real time movies?

Why? Should be *incredibly easy* to get attosecond time resolution:

$$\Delta E \cdot \Delta t \sim \frac{\hbar}{2} \qquad \Delta t \sim 100\, as \;\;\Rightarrow\;\; \Delta E \sim 7\, eV$$

Can we FT to observe a propagator directly?

Answer: No

$$S(\mathbf{q},\omega) = -\frac{1}{\pi}\frac{1}{1-e^{-\hbar\omega/kT}} \boxed{\mathrm{Im}\left[\chi(\mathbf{q},\omega)\right]}$$

Oops

Our information is incomplete.* Cannot Fourier transform with only the imaginary part.

***This is what Fermi called the "inverse scattering" problem.**

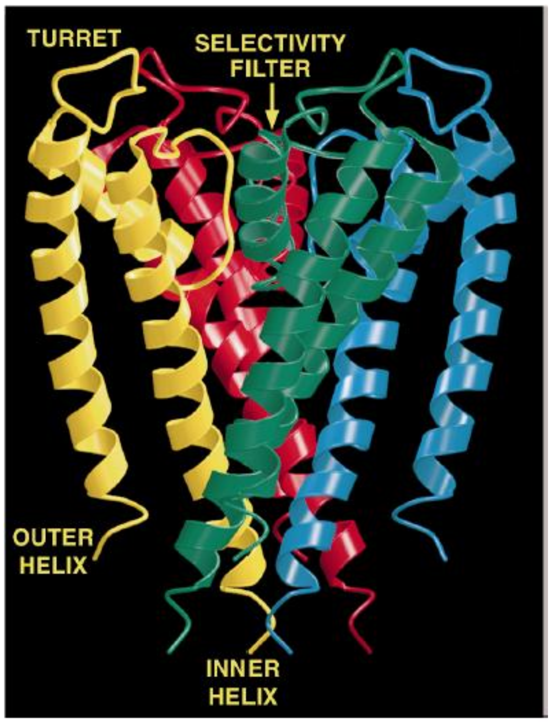
The phase problem reexamined

Central Dogma of x-ray crystallography:

$$I(\mathbf{q}) \propto |\rho(\mathbf{q})|^2$$

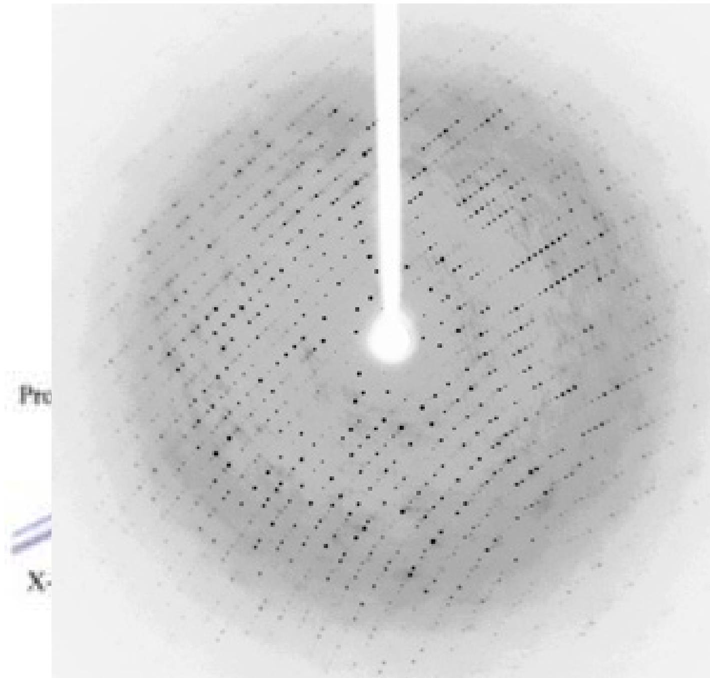
Periodic system (i.e., a crystal):

$$\rho(\mathbf{r}) = \sum_{\mathbf{G}} \rho_{\mathbf{G}} e^{-i\mathbf{G}\cdot\mathbf{r}} \qquad I_{\mathbf{G}} \propto |\rho_{\mathbf{G}}|^2$$



KcsA channel
R. MacKinnon
Chem. Nobel
Prize, 2003

D. A. Doyle, et al., Science **280**, 69 (1998)



- **Phase problem is solved by incorporating *constraints* (Hg or Se atoms)**
- **This is the basis for the field of *structural genomics***
- **Based on *classical scattering* theory. All scattering is *elastic.***

The phase problem reexamined

Van Hove function:

$$S(\mathbf{q},\omega) = \frac{1}{\hbar}\sum_n \left|\left\langle n\right|\hat{\rho}(\mathbf{q})\left|0\right\rangle\right|^2 \delta(\omega-\omega_0+\omega_m) \qquad (\,T=0\,)$$

What we think we measure:

$$S(\mathbf{q},\omega)\Big|_{\omega=0} = \frac{1}{\hbar}\left|\left\langle 0\right|\hat{\rho}(\mathbf{q})\left|0\right\rangle\right|^2 = \frac{1}{\hbar}\left|\left\langle \hat{\rho}(\mathbf{q})\right\rangle\right|^2$$

What we actually measure:

$$\int d\omega\, S(\mathbf{q},\omega) = \frac{1}{\hbar}\sum_n \left|\left\langle n\right|\hat{\rho}(\mathbf{q})\left|0\right\rangle\right|^2 = \frac{1}{\hbar}\left\langle 0\right|\hat{\rho}(-\mathbf{q})\hat{\rho}(\mathbf{q})\left|0\right\rangle = \frac{1}{\hbar}\left\langle \left|\hat{\rho}(\mathbf{q})\right|^2\right\rangle$$

More general formulation of the phase problem:

$$S(\mathbf{q},\omega) = -\frac{1}{\pi}\frac{1}{1-e^{-\hbar\omega/kT}}\mathrm{Im}\left[\chi(\mathbf{q},\omega)\right] \qquad \mathrm{Re}\left[\chi(\mathbf{k},\omega)\right] = \frac{2}{\pi}P\int_0^\infty \frac{\mathrm{Im}\left[\chi(\mathbf{k},\omega')\right]}{(\omega')^2-\omega^2}d\omega'$$

- $\chi(\mathbf{x},t) = 0$ for $t < 0$
- **Raw spectra do not really describe dynamics – no causal information**
- **Causality is the constraint. Must assign an *arrow of time* to the problem.**
- **Rise of entropy ⇔ arrow of time**

What if the system is inhomogeneous?

Assume it's periodic:

$$\chi(\mathbf{x}_1, \mathbf{x}_2, t_1 - t_2) = -\frac{i}{\hbar}\left\langle\left[\hat{\rho}(\mathbf{x}_2, t_2), \hat{\rho}(\mathbf{x}_1, t_1)\right]\right\rangle \theta(t_2 - t_1)$$

$$\chi(\mathbf{k}_1, \mathbf{k}_2, \omega) = \chi(\mathbf{k}_1, \mathbf{G} - \mathbf{k}_1, \omega)$$

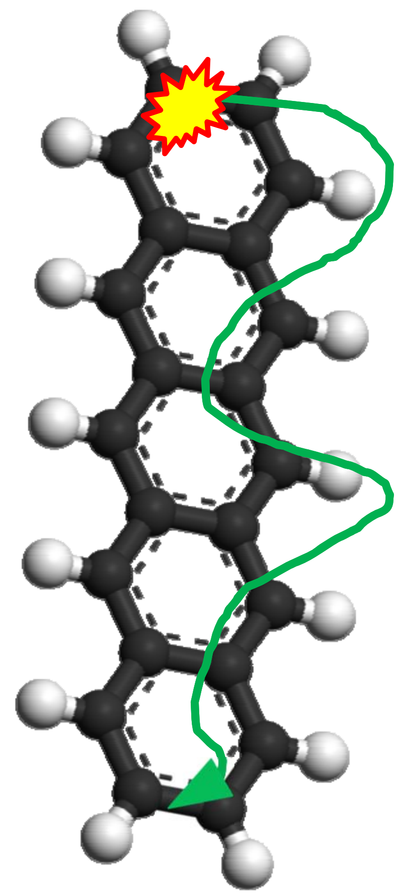
In regular scattering, we only measure the diagonal (G=0) components of this matrix:

$$S(\mathbf{k}, \omega) = -\frac{1}{\pi}\frac{1}{1 - e^{-\hbar\omega/kT}} \mathrm{Im}\left[\chi(\mathbf{k}, -\mathbf{k}, \omega)\right]$$

Naïvely Fourier transform and you get a spatial average:

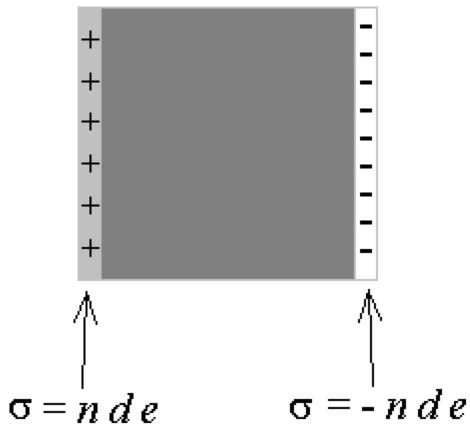
$$\chi(\mathbf{r}, t) = \int d\mathbf{r}' \, \chi(\mathbf{r}', \mathbf{r}' + \mathbf{r}, t)$$

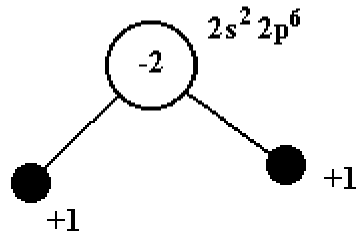
[P. A., et al., Phys. Rev. B **80**, 054302 (2009)]



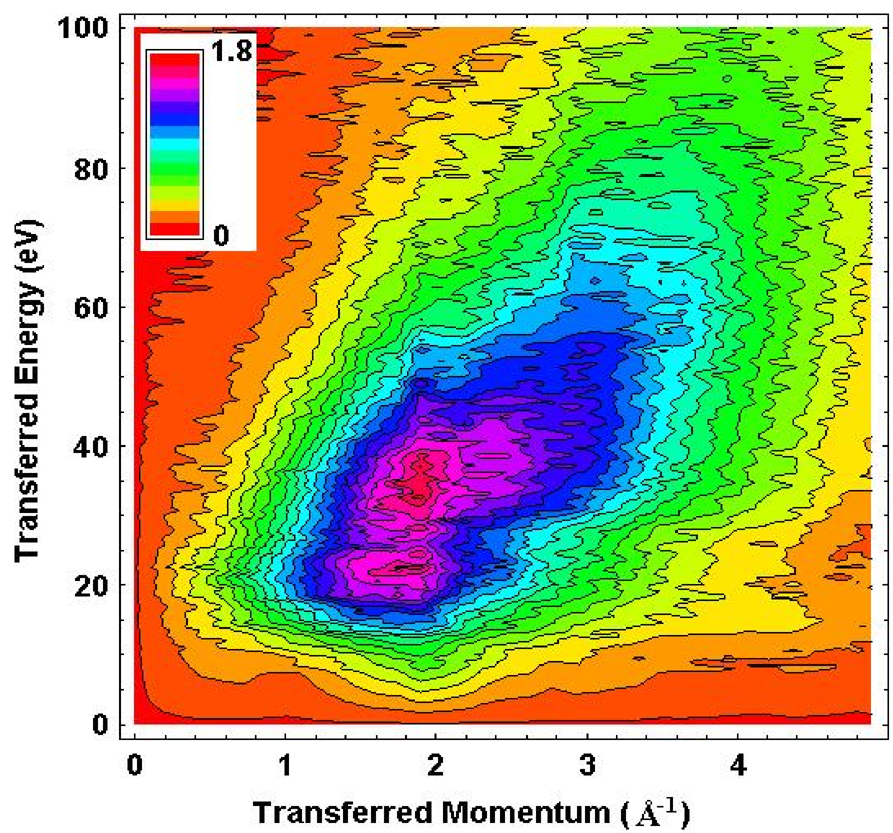
If the system is homogeneous, this is OK. If not, things get even better… but let's start with the homogeneous case.

Plasma oscillations in water





$-\,\mathrm{Im}[\chi(\mathbf{k},\omega)]\quad (as/\text{Å}^3)$



- **8 valence electrons / molecule**
- **ρ = 1 g/cm^3 $\Rightarrow$ n = 0.20 e/Å^3**
- **$\omega_p = \sqrt{(4\pi n e^2/m)}$ = 16.6 eV**

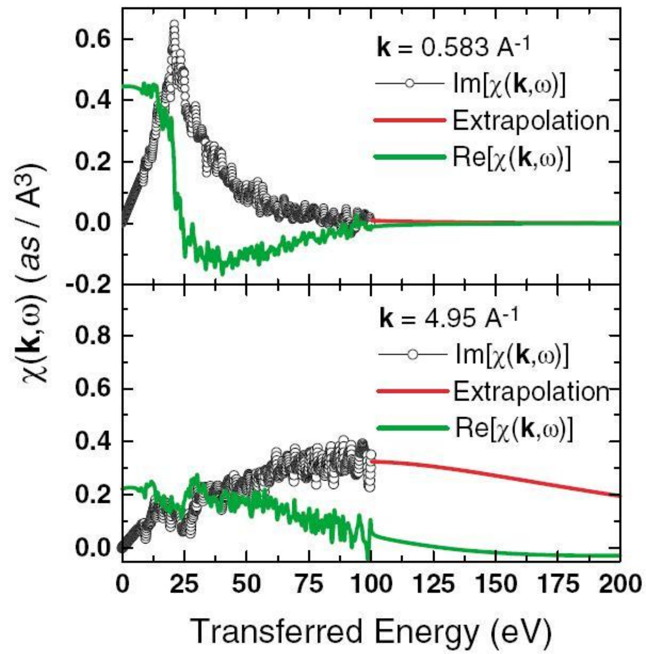
Problems

Problem #1:

Im[χ(**k**,ω)] must be defined on *infinite* ω interval for continuous time interval

Solution:

Extrapolate.



Side effects:

- $\chi(\mathbf{x},t)$ defined on continuous (infinitely narrow) time intervals.
- Time "resolution" $\Delta t_N = \pi/\Omega_{max}$
- Ω_{max} plays role of pulse width.

More Problems

Problem #2:

Discrete points violate causality

Im[χ(**k**,ω)] must be defined on *continuous* ω interval. Periodicity incompatible with causality.

Solution:

Interpolation (i.e., add data)

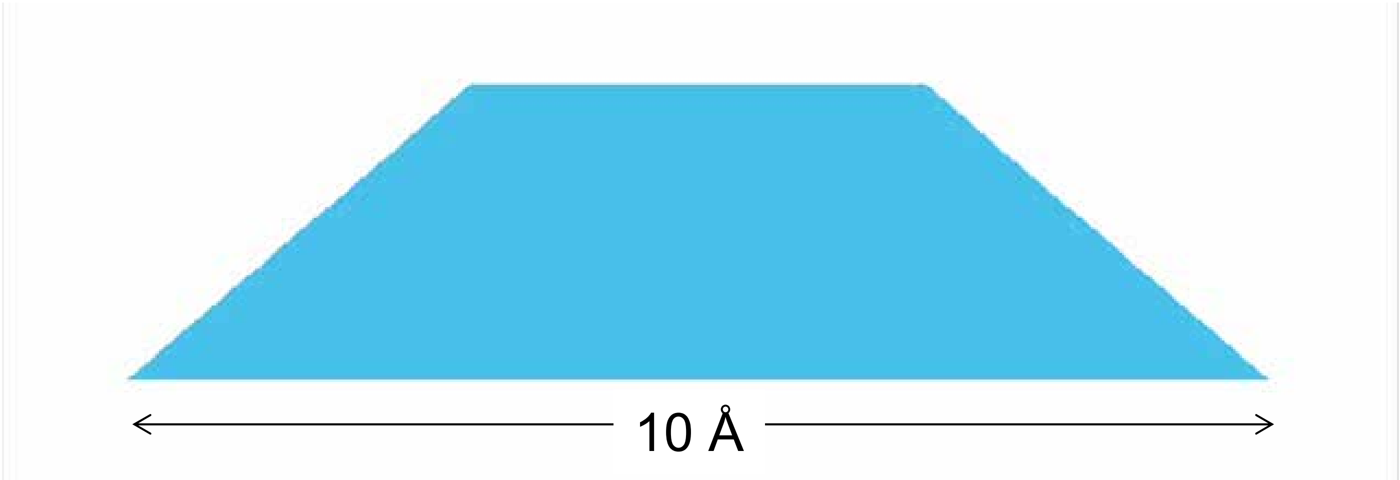
$$\chi(\mathbf{k},t) = \int_0^\infty \frac{d\omega}{\pi}\left[\sin(\omega t)\, Im\chi(\mathbf{k},\omega) \;+\; \cos(\omega t)\, Re\chi(\mathbf{k},\omega)\right]$$

$$\chi(\mathbf{k},t) = \frac{2}{\pi}\int_0^\infty d\omega \sin(\omega t)\, Im\chi(\mathbf{k},\omega)$$

Side effects:

- χ(**x**,t) defined forever. Vanishes for $t < 0$.
- Repeats with period $T = 2\pi/\Delta\omega$
- $\Delta\omega$ plays role of rep rate

Disturbance from a point source in water



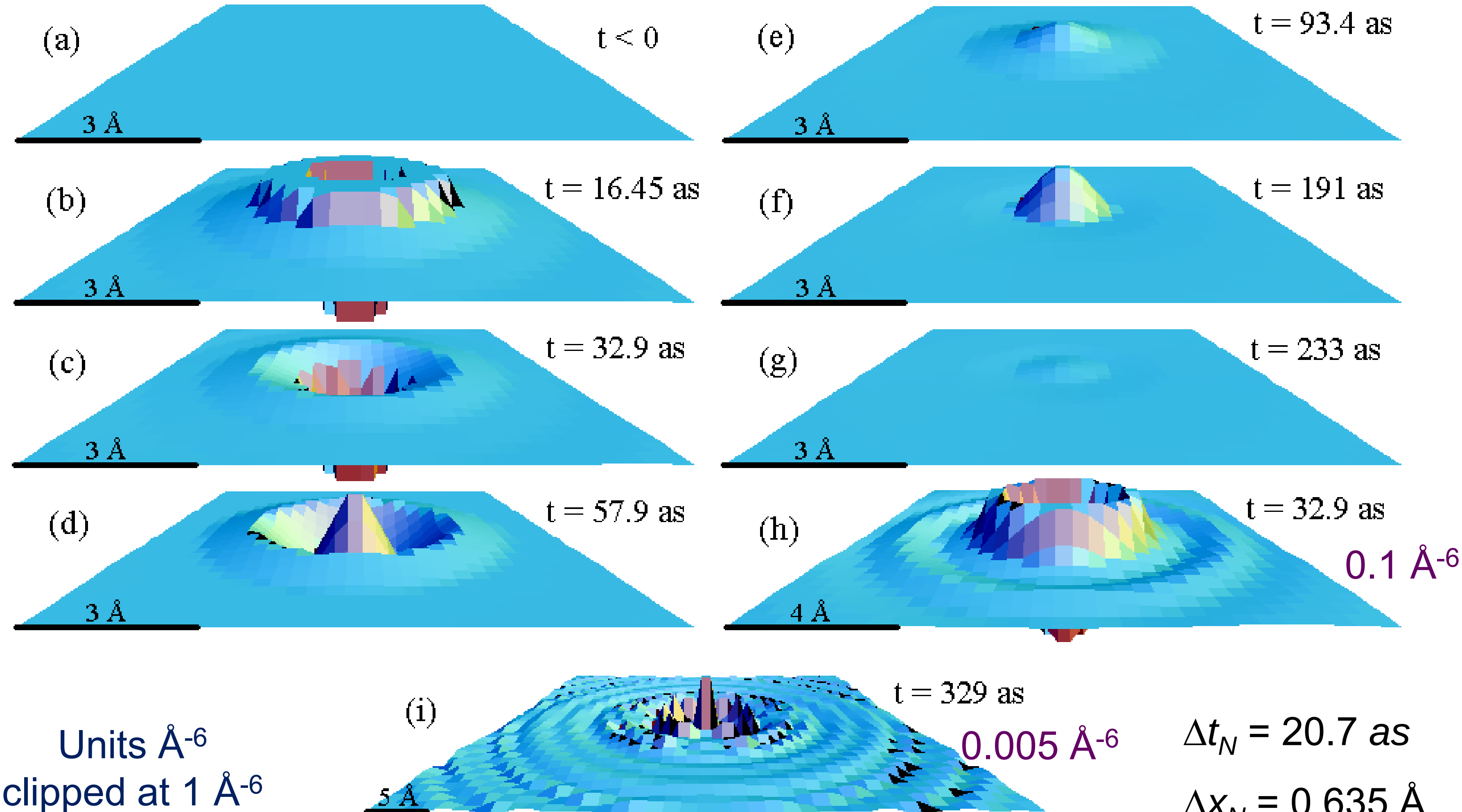
Units Å^{-6}
clipped at 1 Å^{-6}

Δt_N = 20.7 *as*

Δx_N = 0.635 Å

P. A., et al., PRL, **92**, 237401 (2004)

Frame-by-frame



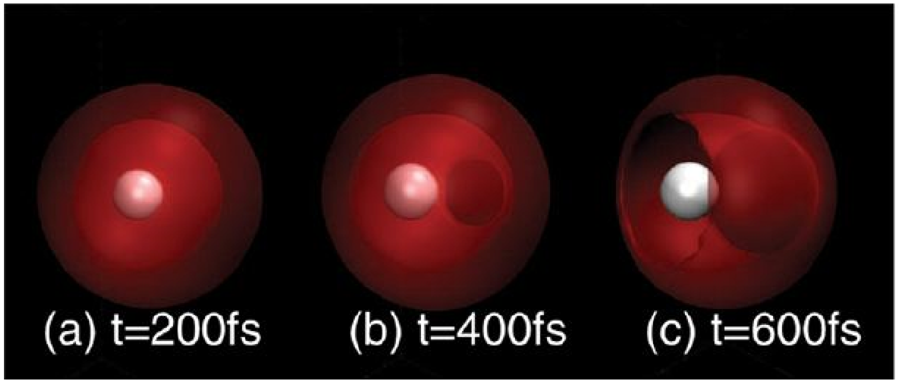
Units Å^{-6}
clipped at 1 Å^{-6}

Δt_N = 20.7 *as*

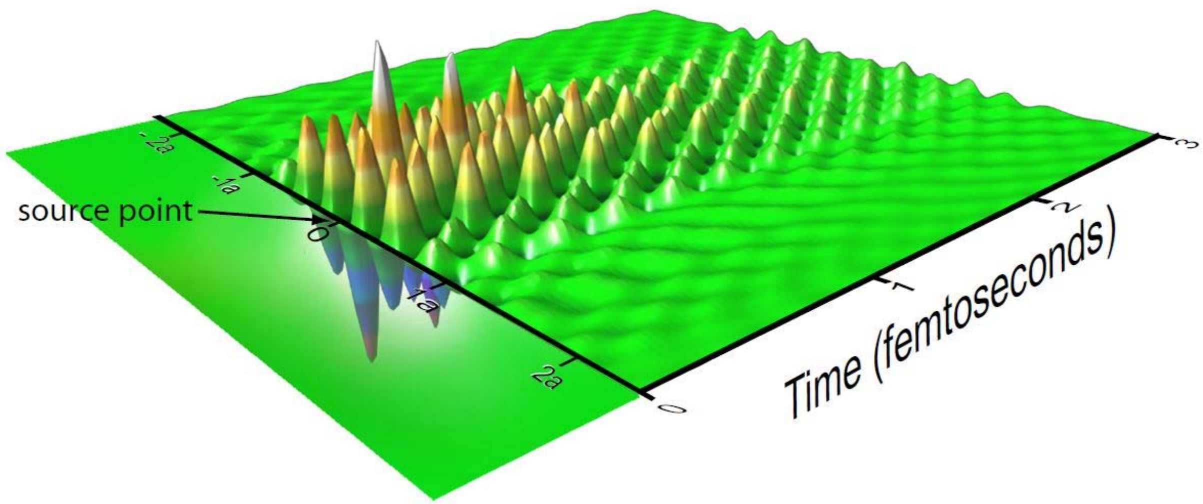
Δx_N = 0.635 Å

- Events transpire in 350 as – *light travels 100 nm in vacuum*
- Causality ⇔ Analytic properties ⇔ Rise of entropy ⇔ Arrow of time

Attosecond imaging with IXS

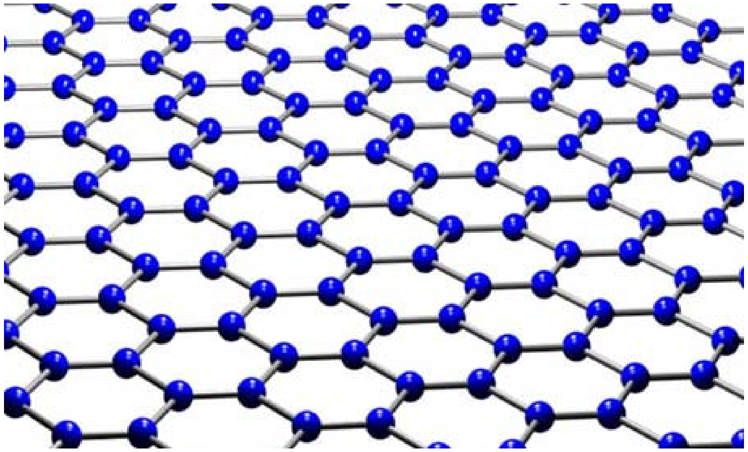


Ion solvation dynamics (Δt = 26 fs)
R. Coridan, et al., PRL **103**, 237402 (2009)



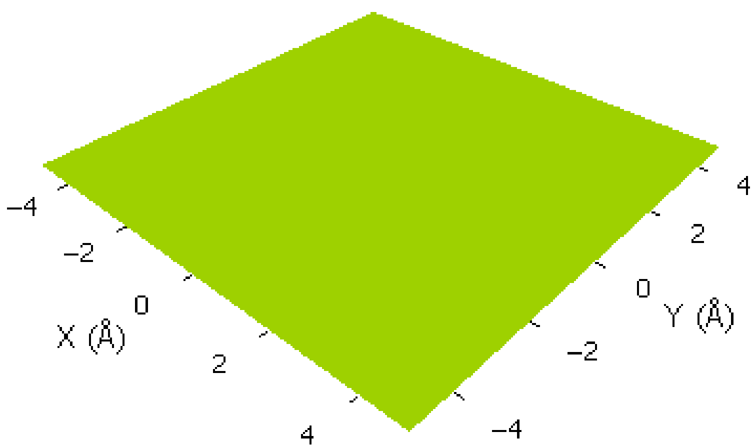
"Birth" of an exciton in LiF (Δt = 20.6 as)
P. A. et al., PNAS **105**, 12159 (2008)

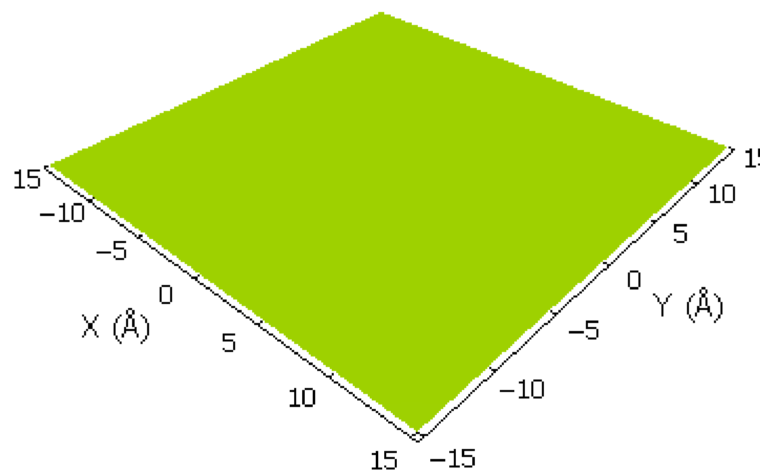
Effective fine structure constant of graphene



$$\alpha_g^*(\mathbf{k},\omega)=\alpha_g\left[1+V(k)\chi(\mathbf{k},\omega)\right]$$

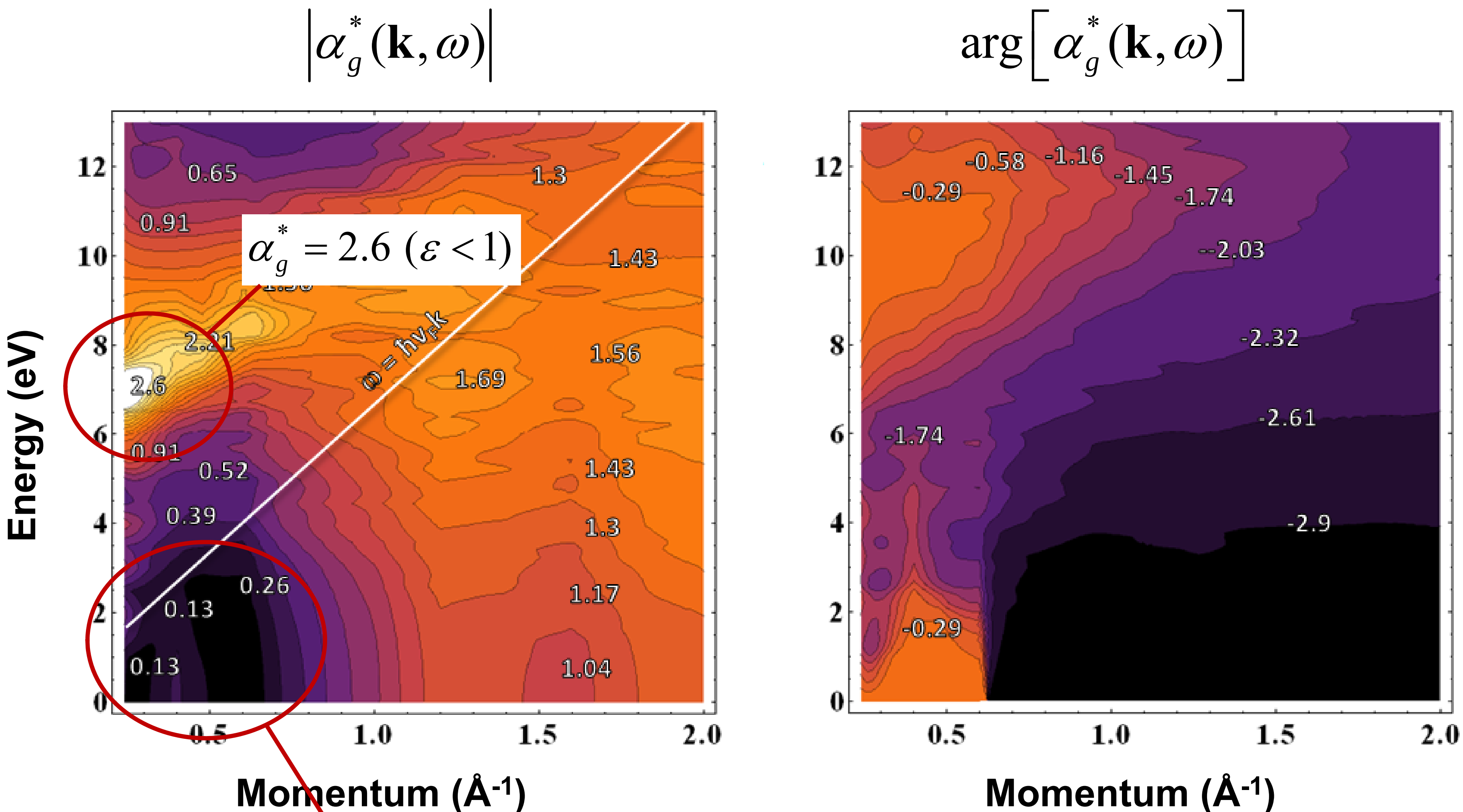
Charge propagator. Gives *screening correction* to α_g.





Δt = 10.3 attoseconds (10^{-17} sec), Δr = 0.2 Å

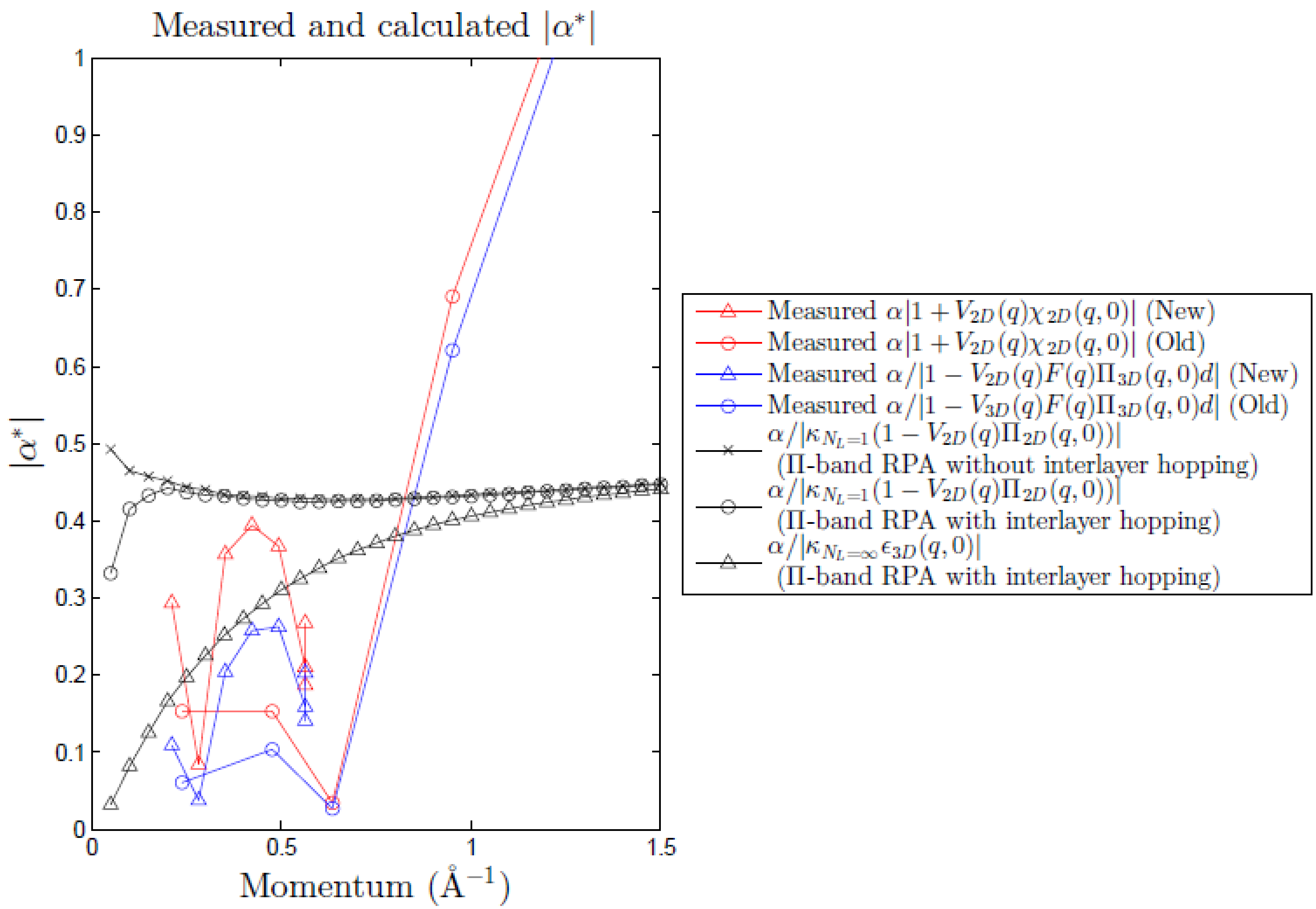
Effective fine structure constant, $\alpha_g^*(\mathbf{k},\omega)$



$$\alpha_g^*(0^+,0) \equiv \lim_{\mathbf{k}\to 0} \alpha_g^*(\mathbf{k},0) = 0.14 \pm 0.092 \approx 1/7$$

J. P. Reed, et al., Science **330**, 805 (2010)

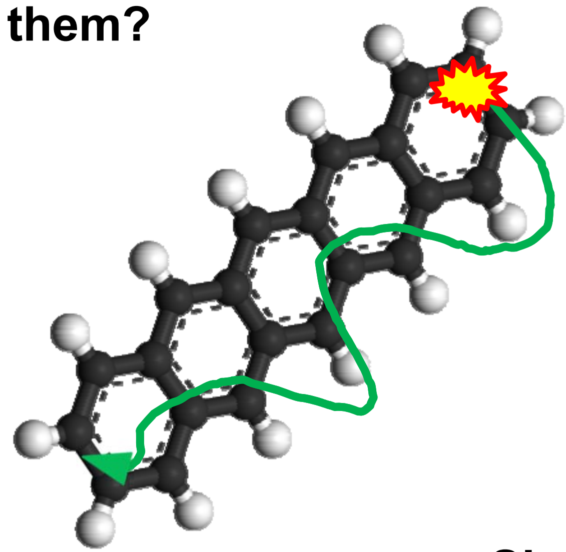
New results



Measured and calculated $|\alpha^*|$
$|\alpha^*|$
Momentum ($\mathrm{\AA}^{-1}$)
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
0
0.5
1
1.5
Measured $\alpha|1 + V_{2D}(q)\chi_{2D}(q, 0)|$ (New)
Measured $\alpha|1 + V_{2D}(q)\chi_{2D}(q, 0)|$ (Old)
Measured $\alpha/|1 - V_{2D}(q)F(q)\Pi_{3D}(q, 0)d|$ (New)
Measured $\alpha/|1 - V_{3D}(q)F(q)\Pi_{3D}(q, 0)d|$ (Old)
$\alpha/|\kappa_{N_L=1}(1 - V_{2D}(q)\Pi_{2D}(q, 0))|$
(Π-band RPA without interlayer hopping)
$\alpha/|\kappa_{N_L=1}(1 - V_{2D}(q)\Pi_{2D}(q, 0))|$
(Π-band RPA with interlayer hopping)
$\alpha/|\kappa_{N_L=\infty}\epsilon_{3D}(q, 0)|$
(Π-band RPA with interlayer hopping)

X-Ray Scattering, Finally Done “Correctly”

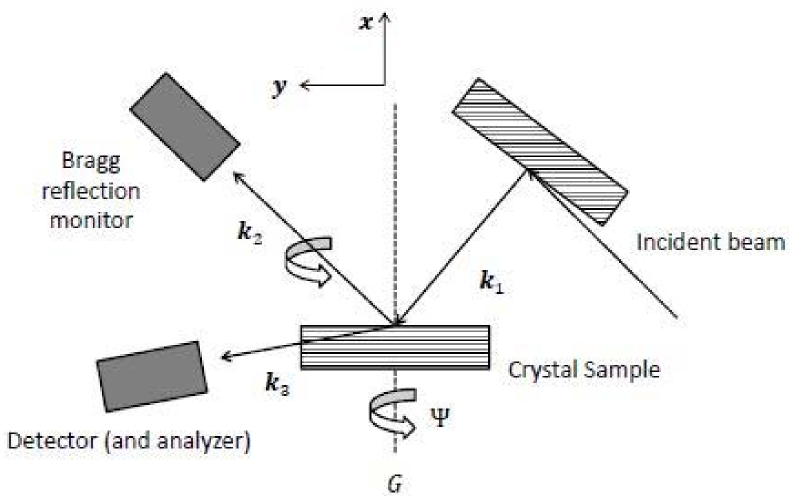
We need the off-diagonal terms. Can we measure them?

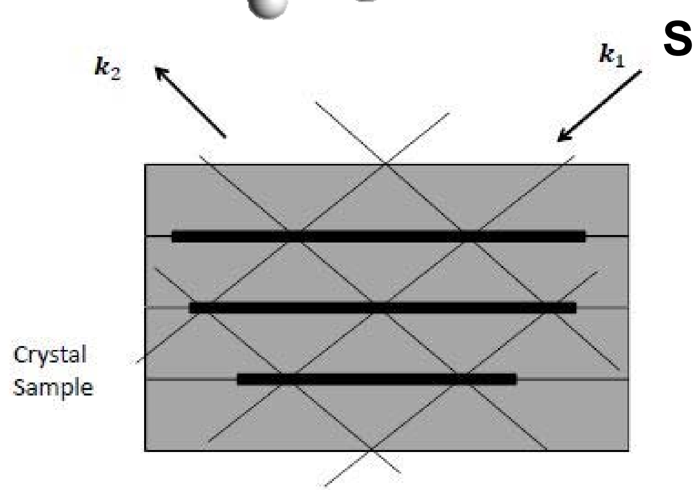


$$\chi(\mathbf{x}_1,\mathbf{x}_2,t_1-t_2) = -\frac{i}{\hbar}\left\langle\left[\hat{\rho}(\mathbf{x}_2,t_2),\hat{\rho}(\mathbf{x}_1,t_1)\right]\right\rangle\theta(t_2-t_1)$$

$$\chi(\mathbf{k}_1,\mathbf{k}_2,\omega) = \chi(\mathbf{k}_1,\mathbf{G}-\mathbf{k}_1,\omega)$$ **(periodic case)**

Yes … X-ray standing waves:





J. A. Golovchenko, et al., PRL 46, 1454 (1981)
W. Schulke, U. Bonse, S. Mourikis, PRL 47, 1209 (1981)
W. Schulke, A. Kaprolat, PRL 67, 879 (1991)

X-Ray Scattering, Finally Done "Correctly"

"Incident" photon is in a superposition of momenta:

$$|i\rangle = (g_1 a^\dagger_{k_1\alpha_1} + g_2 a^\dagger_{k_2\alpha_2} e^{i\gamma})\,|m\rangle\,. \qquad\qquad |f\rangle = a^\dagger_{k_3\alpha_3}\,|n\rangle$$

Now do scattering:

$$\hat{H} = \hat{H}_0 + \frac{e}{2mc}\int \hat{\psi}^\dagger \hat{\mathbf{A}}\cdot\boldsymbol{p}\,\hat{\psi}\,d\boldsymbol{x} + \frac{e^2}{2mc^2}\int \hat{\rho}\,\hat{\mathbf{A}}^2\,d\boldsymbol{x}$$

The cross section contains interference terms:

$$\begin{aligned}\frac{d^2\sigma}{dE'd\Omega} = \left(\frac{e^2}{mc^2}\right)^2 \frac{\omega_3}{\omega_1}\,\big[& g_1^2|\hat{\boldsymbol{\epsilon}}_3^*\cdot\hat{\boldsymbol{\epsilon}}_1|^2 S(\boldsymbol{q}_1,\omega) + g_2^2|\hat{\boldsymbol{\epsilon}}_3^*\cdot\hat{\boldsymbol{\epsilon}}_2|^2 S(\boldsymbol{q}_2,\omega) \\ & + g_1 g_2 e^{i\gamma}(\hat{\boldsymbol{\epsilon}}_3\cdot\hat{\boldsymbol{\epsilon}}_1^*)(\hat{\boldsymbol{\epsilon}}_3^*\cdot\hat{\boldsymbol{\epsilon}}_2) S(\boldsymbol{q}_1,\boldsymbol{q}_2,\omega) \\ & + g_1 g_2 e^{-i\gamma}(\hat{\boldsymbol{\epsilon}}_3^*\cdot\hat{\boldsymbol{\epsilon}}_1)(\hat{\boldsymbol{\epsilon}}_3\cdot\hat{\boldsymbol{\epsilon}}_2^*) S(\boldsymbol{q}_2,\boldsymbol{q}_1,\omega)\big]\,,\end{aligned}$$

Phase of the standing wave. ***Requires coherent wave field.***

Interference terms are off-diagonal dynamic structure factor

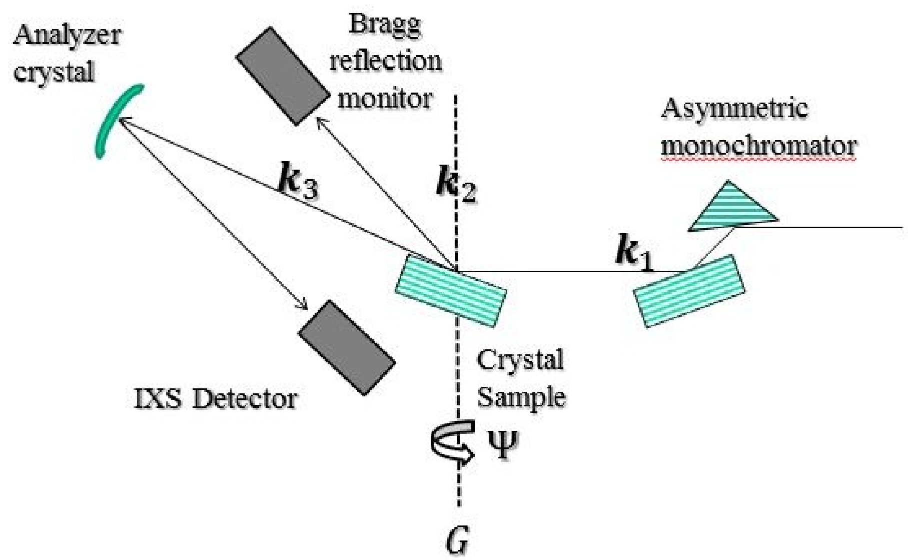
$$S(\boldsymbol{q}_1,\boldsymbol{q}_2,\omega) \equiv \sum_{n,m} b_m \langle m|\,\rho(\boldsymbol{q}_1)\,|n\rangle\,\langle n|\,\rho(-\boldsymbol{q}_2)\,|m\rangle\,\delta(\hbar\omega - E_n + E_m)$$

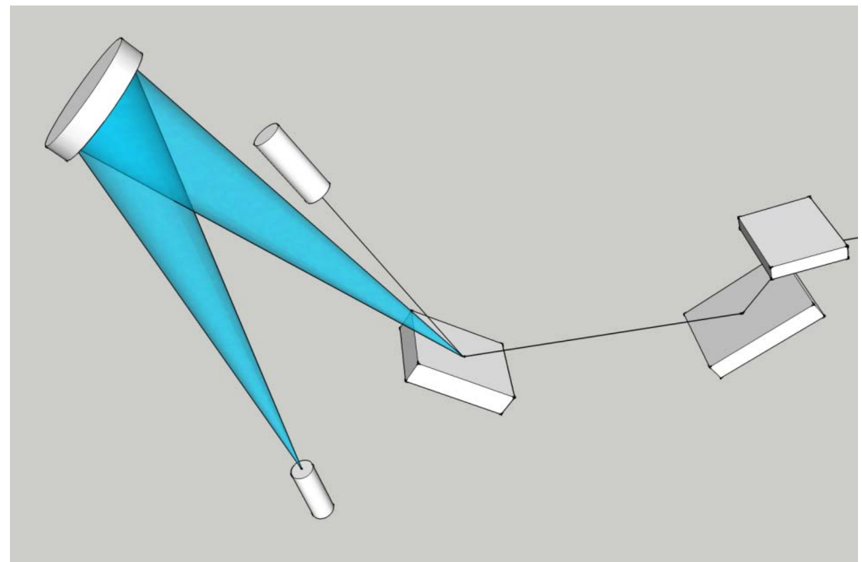
Generalized fluctuation-dissipation theorem:

$$S(\boldsymbol{q}_1,\boldsymbol{q}_2,\omega) = -\frac{1}{\pi}\frac{1}{1-e^{-\beta\hbar\omega}}\,\mathrm{Im}\,\chi(\boldsymbol{q}_1,-\boldsymbol{q}_2,\omega).$$

Y. Gan, A. Kogar, P. Abbamonte, Chem. Phys. 414, 160 (2012)

“Crystallography for the collective excitations”

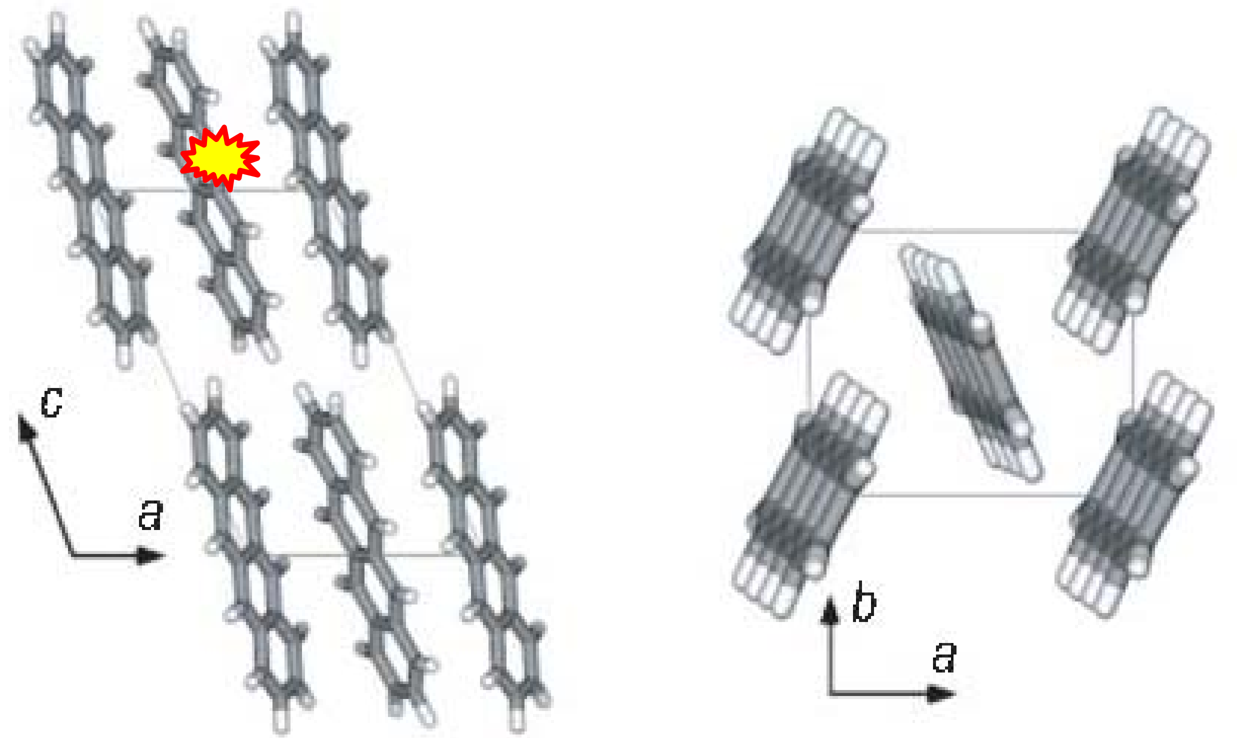




Y. Gan, A. Kogar, P. Abbamonte, Chem. Phys. 414, 160 (2012)

"Crystallography for the collective excitations"

- **Complete reconstruction of** $\chi(\mathbf{r}_1, \mathbf{r}_2, t)$
- **Angstrom spatial resolution**
- **Attosecond time resolution**



Y. Gan, A. Kogar, P. Abbamonte, Chem. Phys. 414, 160 (2012)

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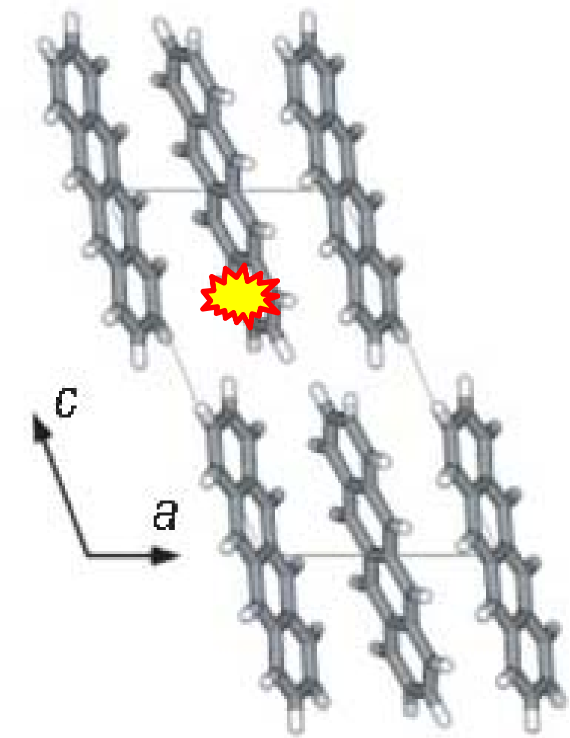

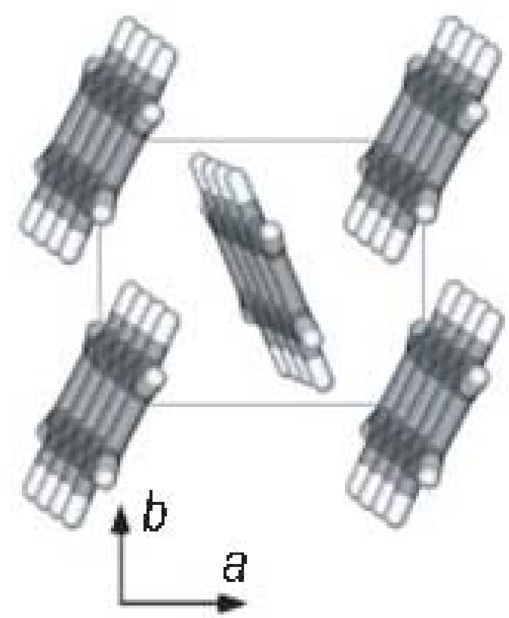

Y. Gan, A. Kogar, P. Abbamonte, Chem. Phys. 414, 160 (2012)