Real-time approaches to x-ray and optical spectra

J. J. Rehr, F. Vila, J. Kas

Department of Physics
University of Washington
Seattle, WA USA

UNIVERSITY OF WASHINGTON

Real-time Approaches for Optical and X-ray Spectra

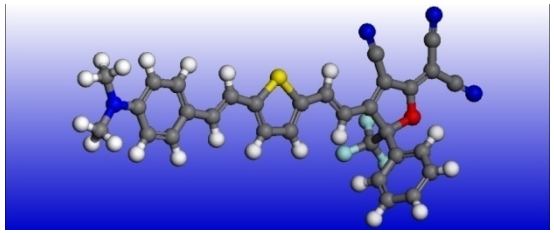
Goal: Real-time linear and non-linear response

Talk:

- **I**. Linear and Nonlinear Optical Response RT-TDDFT
- **II.** Real-time core-level XAS
- **III.** Many-body Effects

I. Real-Space & Real-Time Linear and Non-linear Optical Response

- Difficulty: frequency-space is computationally demanding **too-many** excited states



- Solution: RT-TDDFT - extension of SIESTA*

*Sanchez-Portal, Tsolakidis, and Martin, Phys. Rev. B**66**, 235416 (2002)

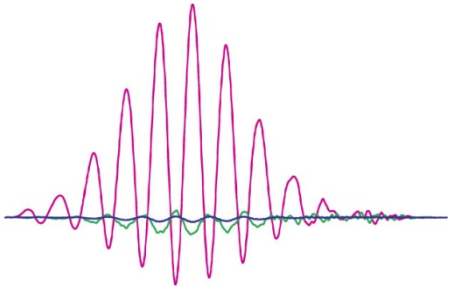
Real-Time-TDDFT*

THE JOURNAL OF CHEMICAL PHYSICS **127**, 154114 (2007)

Real-time time-dependent density functional theory approach for frequency-dependent nonlinear optical response in photonic molecules

Y. Takimoto, F. D. Vila, and J. J. Rehr[a)]
Department of Physics, University of Washington, Seattle, Washington 98195, USA

(Received 11 July 2007; accepted 4 September 2007; published online 19 October 2007)



We present *ab initio* calculations of frequency-dependent linear and nonlinear optical responses based on real-time time-dependent density functional theory for arbitrary photonic molecules. This approach is based on an extension of an approach previously implemented for a linear response using the electronic structure program SIESTA. Instead of calculating excited quantum states, which can be a bottleneck in frequency-space calculations, the response of large molecular systems to time-varying electric fields is calculated in real time. This method is based on the finite field approach generalized to the dynamic case. To speed the nonlinear calculations, our approach uses Gaussian enveloped quasimonochromatic external fields. We thereby obtain the frequency-dependent second harmonic generation $\beta(-2\omega;\omega,\omega)$, the dc nonlinear rectification $\beta(0;-\omega,\omega)$, and the electro-optic effect $\beta(-\omega;\omega,0)$. The method is applied to nanoscale photonic nonlinear optical molecules, including *p*-nitroaniline and the FTC chromophore, i.e., 2-[3-Cyano-4-(2-{5-[2-(4-diethylamino-phenyl)-vinyl] - thiophen-2-yl} - vinyl)-5,5-dimethyl-5H-furan-2-ylidene]-malononitrile, and yields results in good agreement with experiment. © *2007 American Institute of Physics*.

*K. Yabana and G. F. Bertsch, Phys. Rev. B **54**, 4484 1996.

Real time Linear Response

$$\delta \mathbf{p}(t) = \mathbf{p}(t) - \vec{\mu}_0$$

Induced Dipole Moment

$$\delta p_i(t) = \int dt' \, \chi_{ij}^{(1)}(t - t') E_j(t')$$

$$\chi_{ij}^{(1)}(\omega) = \delta p_i(\omega)/E_j(\omega) = \alpha_{ij}(\omega)$$

Linear Response Function

$$\epsilon_{ij}(\omega) = 1 + 4\pi N \alpha_{ij}(\omega)$$

Linear Dielectric Function

$$\sigma(\omega) \sim \omega \, \langle \alpha(\omega) \rangle / E(\omega)$$

Optical Absorption

RT-TDDFT Formalism

- **Yabana and Bertsch** Phys. Rev. B**54**, 4484 (1996)

$$i\frac{\partial \Psi}{\partial t} = H(t)\,\Psi \qquad H = -\frac{1}{2}\nabla^2 + V_{ext}(\mathbf{r},t) + V_H[\rho](\mathbf{r},t) + V_{xc}[\rho](\mathbf{r},t)$$

- Direct numerical integration of TD Kohn-Sham equations

$$\Psi(t) = T\,\exp\left(-i\int_0^t H(t')dt'\right)\Psi(0)$$

- The response to external field is determined by applying a **time-dependent electric field** $\Delta H(t) = -\mathbf{E}(t)\cdot\mathbf{x}$.

- Optical properties determined from **total dipole moment**:

$$\mathbf{p}(t) = \int \rho(\mathbf{r},t)\,\mathbf{r}\,d^3\mathbf{r}$$

Sometimes more EFFICIENT than Frequency space methods

Numerical Real-time Evolution

- Ground state density ρ_0, **overlap matrix** $\boldsymbol{S}$**,** and $\boldsymbol{H}(t)$ at each time-step evaluated with SIESTA

$$i\frac{\partial c(t)}{\partial t} = S^{-1}H(t)c(t)$$

Coefficients of Orbitals

- Crank-Nicholson time-evolution: **unitary**, **time-reversible**

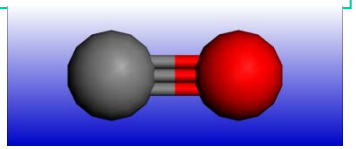
Stable for long time-steps !

$$c(t+\Delta t) = \frac{1 - iS^{-1}H(\bar{t})\Delta t/2}{1 + iS^{-1}H(\bar{t})\Delta t/2}c(t) + \mathcal{O}(\Delta t^2), \quad \bar{t} = t + \Delta t/2$$

- Adiabatic GGA exchange-correlation (PBE) functional

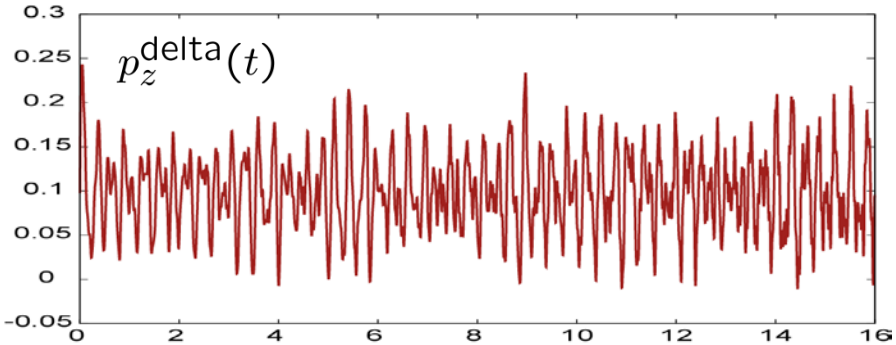
Example: CO *Linear Response*

$p_z(t)$ response due to applied $E_z(t)$

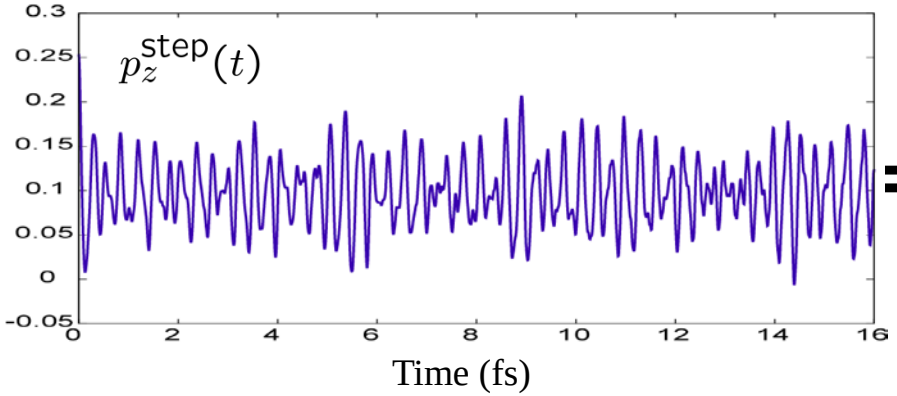


- Delta Function

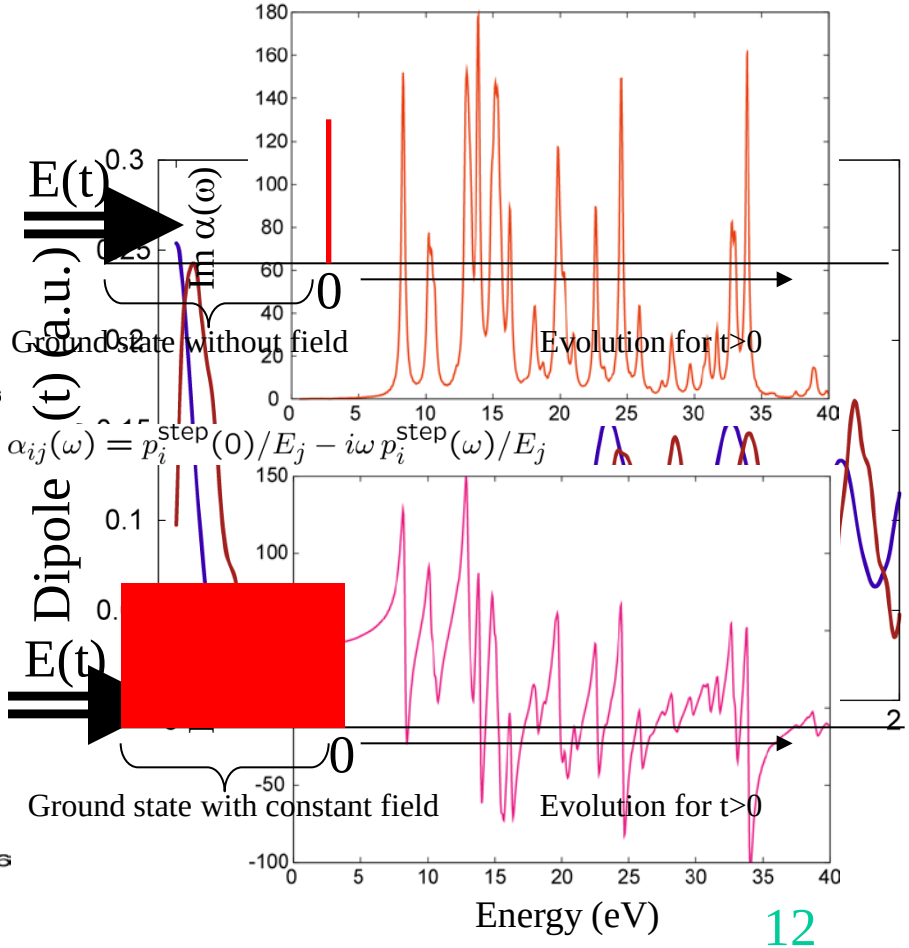
  (Unit Impulse at *t=0*)



(Turn-off Constant E at *t=0*)



$$\alpha_{ij}(\omega) = p_i^{\text{delta}}(\omega)/E_j$$



$$\alpha_{ij}(\omega) = p_i^{\text{step}}(0)/E_j - i\omega\, p_i^{\text{step}}(\omega)/E_j$$

Real time Nonlinear Response

- The nonlinear expansion in field strength

- Accounting for time lag in system response

$$P = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots$$

$$\begin{aligned} p_i(t) = \quad & \mu_i^0 + \int dt_1\, \chi_{ij}^{(1)}(t-t_1) E_j(t_1) \\ & + \int dt_1 \int dt_2\, \chi_{ijk}^{(2)}(t-t_1, t-t_2) E_j(t_1) E_k(t_2) \\ & + \int dt_1 \int dt_2 \int dt_3\, \chi_{ijkl}^{(3)}(t-t_1, t-t_2, t-t_3) E_j(t_1) E_k(t_2) E_l(t_3) \\ & + \cdots \end{aligned}$$

¿ How can we invert the equation to get nonlinear response function?

Extraction of Dynamic Nonlinear Polarizabilities

- Set $E_j(t) = F(t)E_j$, and define expansion $p_i(E)$

$$p_i(t) = \mu_i^0 + p_{ij}^{(1)}(t)E_j + p_{ijk}^{(2)}(t)E_jE_k + \cdots$$

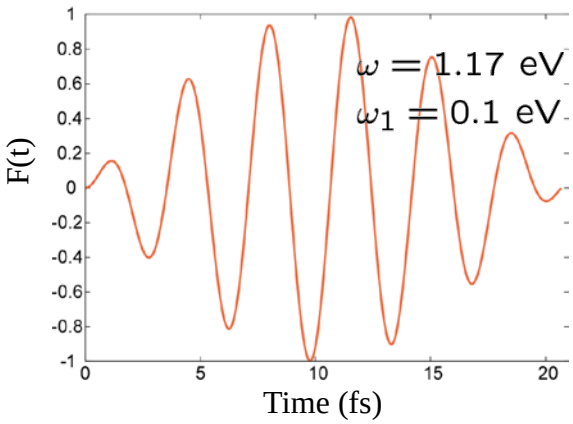
where $p^{(1)}$ yields linear response, $p^{(2)}$ first non-linear quadratic response, ….

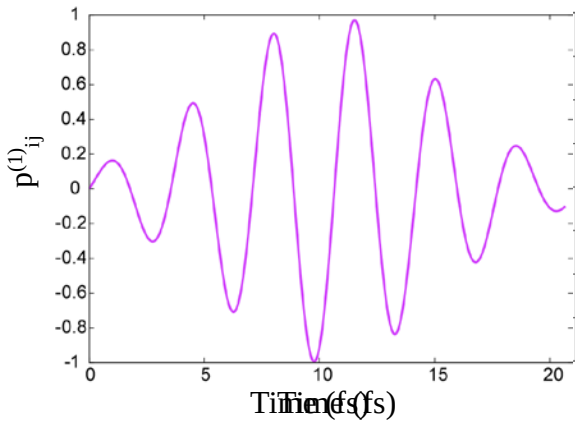
- Quadratic response $\chi^{(2)}$ is then

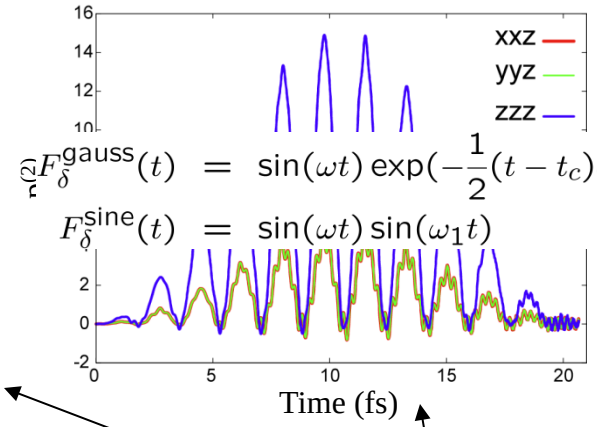
$$p_{ijk}^{(2)}(t) = \int dt_1 \int dt_2 \; \chi_{ijk}^{(2)}(t-t_1, t-t_2)F(t_1)F(t_2)$$

Dynamic Nonlinear Response with Quasi-monochromatic Field $F_\delta(t)$

- Sine wave enveloped by another sine wave or Gaussian

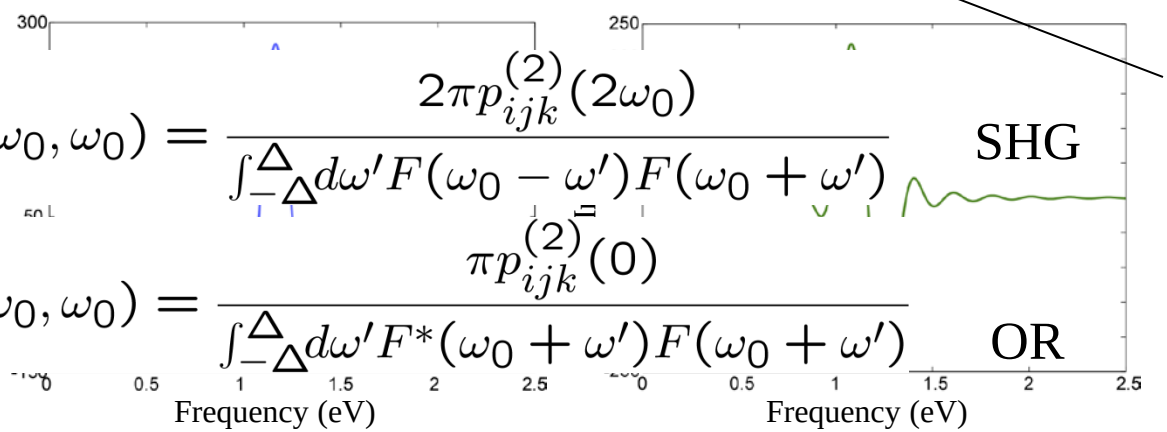






$$F_\delta^{\text{gauss}}(t) = \sin(\omega t)\exp(-\frac{1}{2}(t-t_c)\delta)$$

$$F_\delta^{\text{sine}}(t) = \sin(\omega t)\sin(\omega_1 t)$$

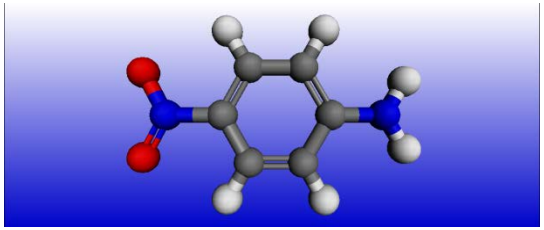


$$\chi^{(2)}_{ijk}(-2\omega_0;\omega_0,\omega_0) = \frac{2\pi p^{(2)}_{ijk}(2\omega_0)}{\int_{-\Delta}^{\Delta} d\omega' F(\omega_0-\omega')F(\omega_0+\omega')}$$ SHG

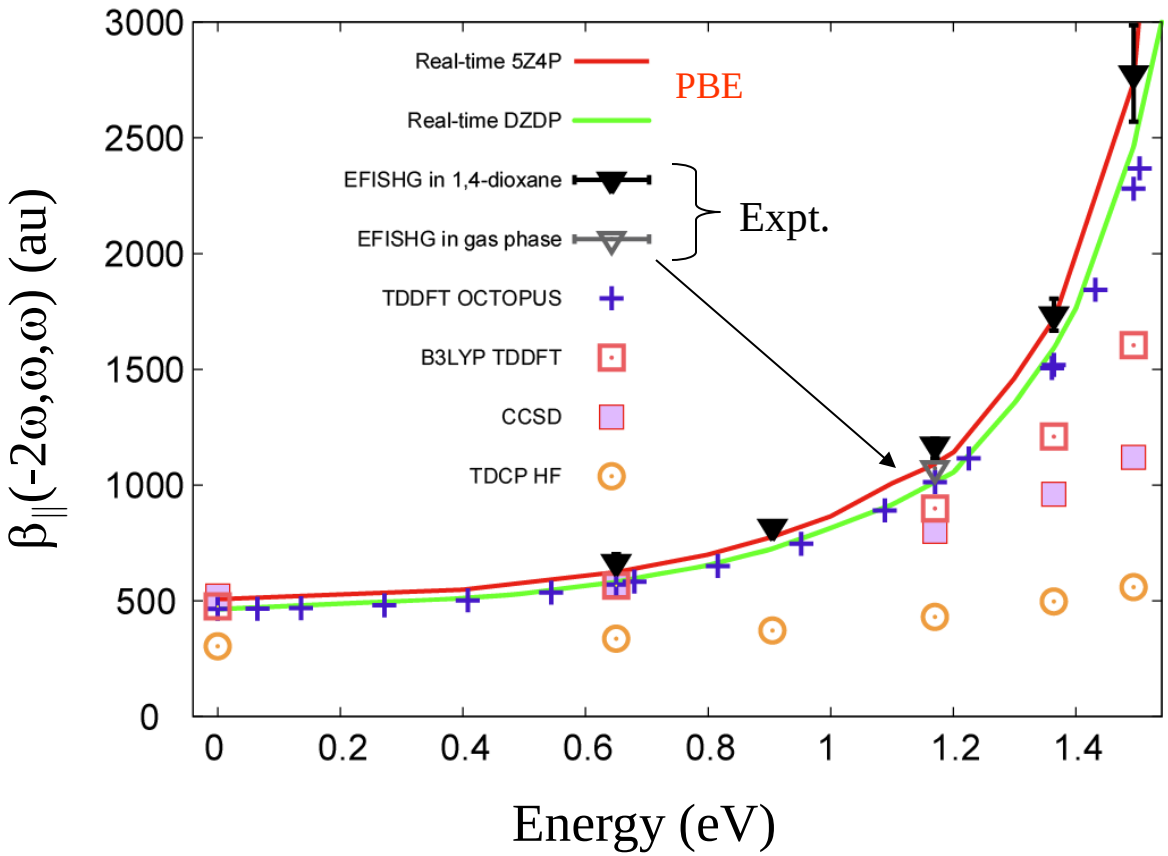
$$\chi^{(2)}_{ijk}(0;-\omega_0,\omega_0) = \frac{\pi p^{(2)}_{ijk}(0)}{\int_{-\Delta}^{\Delta} d\omega' F^*(\omega_0+\omega')F(\omega_0+\omega')}$$ OR

Linear and Nonlinear response of CO

Example pNA: *Nonlinear SHG*



- Comparison with other methods



II. Real-time core-level XAS

Time-correlation function approach

PHYSICAL REVIEW B **86**, 115107 (2012)

Local time-correlation approach for calculations of x-ray spectra

A. J. Lee, F. D. Vila, and J. J. Rehr
Department of Physics, University of Washington, Seattle, Washington 98195, USA
(Received 26 June 2012; published 7 September 2012)

We present a local time-correlation function method for real-time calculations of core level x-ray spectra (RTXS). The approach is implemented in a local orbital basis using a Crank-Nicolson time-evolution algorithm applied to an extension of the SIESTA code, together with projector augmented wave (PAW) atomic transition matrix elements. Our RTXS is formally equivalent to ΔSCF (Δ self consistent field) Fermi's golden rule calculations with a screened core-hole and an effective independent particle approximation. Illustrative calculations are presented for several molecular and condensed matter systems and found to be in good agreement with experiment. The method can also be advantageous compared to conventional frequency-space methods.

$$\mu(\omega) = \frac{1}{\pi}\mathrm{Re}\int_0^\infty dt\, e^{i\omega t} G_c(t)\langle\psi(t)|\psi(0)\rangle\theta(\omega+\epsilon_c-E_F). \qquad (1)$$

$$|\psi(0)\rangle = d|b\rangle$$

XANES with time-dependent DFT

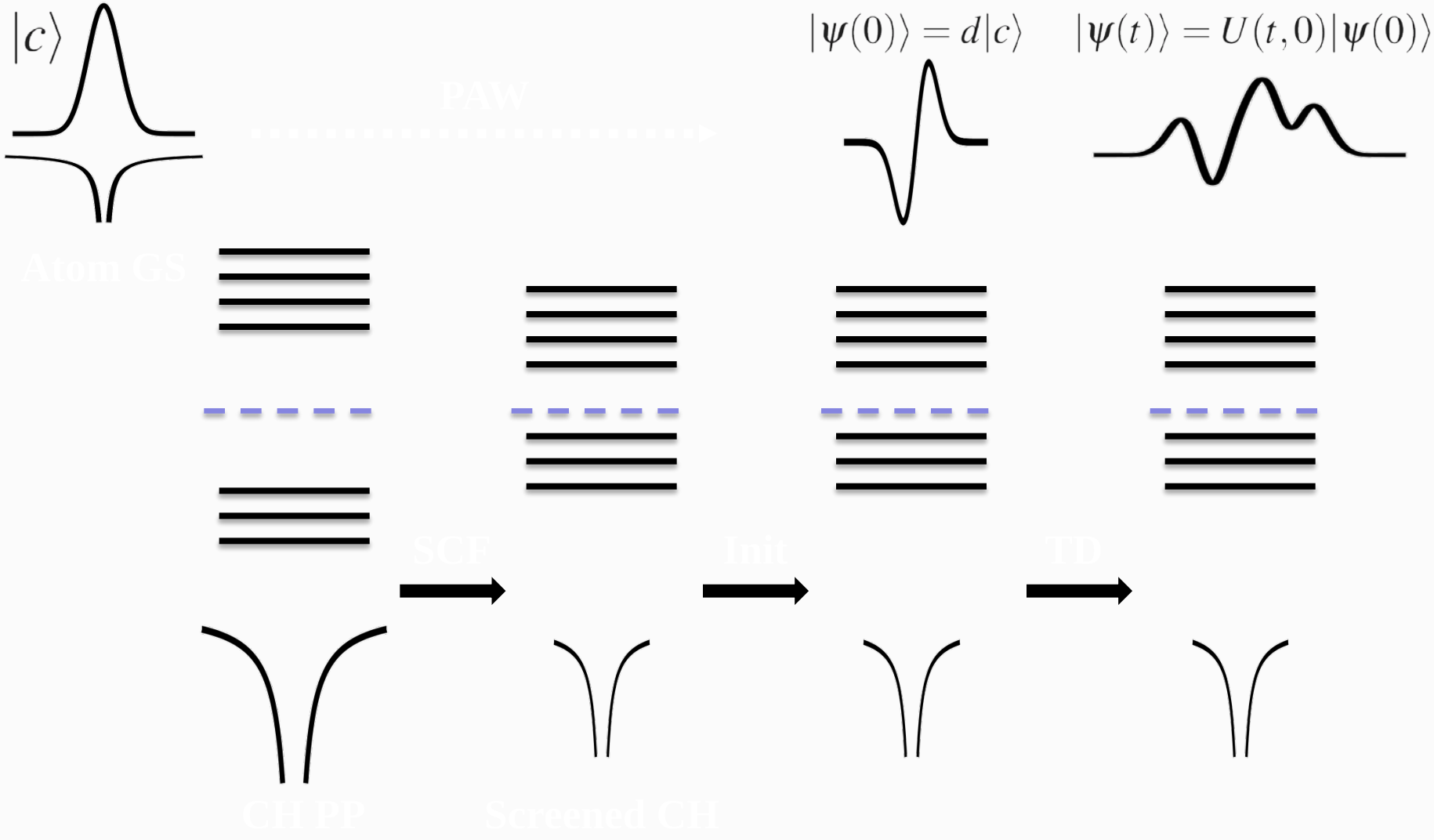
Goal:

Time-dependent x-ray response
Include core hole dynamics

Why use a real-time approach?

New experimental pulsed sources (XFEL, LCLS)
Pump-probe experiments
Increased interest in time-dependent (TD) response

RTXS: The cartoon view



RTXS equations

XAS Absorption (FGR, ΔSCF, FSR)

$$\mu(\omega) = \sum_k |\langle c|d|k\rangle|^2 \delta_\Gamma(\omega + \varepsilon_c - \varepsilon_k)\theta(E - E_F)$$

FT

$$\mu(\omega) = \frac{1}{\pi}\mathrm{Re}\int_0^\infty dt\, e^{i\omega t} G_c(t)\langle\psi(t)|\psi(0)\rangle\theta(\omega + \varepsilon_c - E_F)$$

Core Hole Green's Function

$$G_c(t) = i\exp[i(\varepsilon_c + i\Gamma)t]$$

Autocorrelation Function

$$\langle\psi(t)|\psi(0)\rangle = \sum_{jj'}\langle c|d^\dagger|j\rangle U_{jj'}(t,0)\langle\tilde{j}'|d|c\rangle$$

$$\langle\psi(0)|\psi(t)\rangle \begin{cases} |\psi(0)\rangle = d|c\rangle \\ |\psi(t)\rangle = U(t,0)|\psi(0)\rangle \end{cases}$$

$$U(t,0) = T\exp\left[-i\int_0^t dt\, H(t)\right]$$

$$|\psi(t)\rangle = \sum_j |j\rangle c_j(t)$$

$$H_{jj'} = \langle j|h_H + v_{ch} + \Sigma|j'\rangle$$

Crank-Nicolson

$$\bar{t} = t + \Delta/2 \qquad \mathbf{U}(t+\Delta,t) = \frac{\mathbf{1} - \mathbf{S}^{-1}\mathbf{H}(\bar{t})\Delta/2}{\mathbf{1} + \mathbf{S}^{-1}\mathbf{H}(\bar{t})\Delta/2}$$

Physical interpretation

Projected density of states p-DOS given by autocorrelation function for Seed state of p-symmetry:

$$\rho_\psi(\omega) = -\frac{1}{\pi|\psi|^2|}\mathrm{Im}\int_0^\infty dt\, e^{i\omega t}\langle\psi(t)|\psi(0)\rangle$$

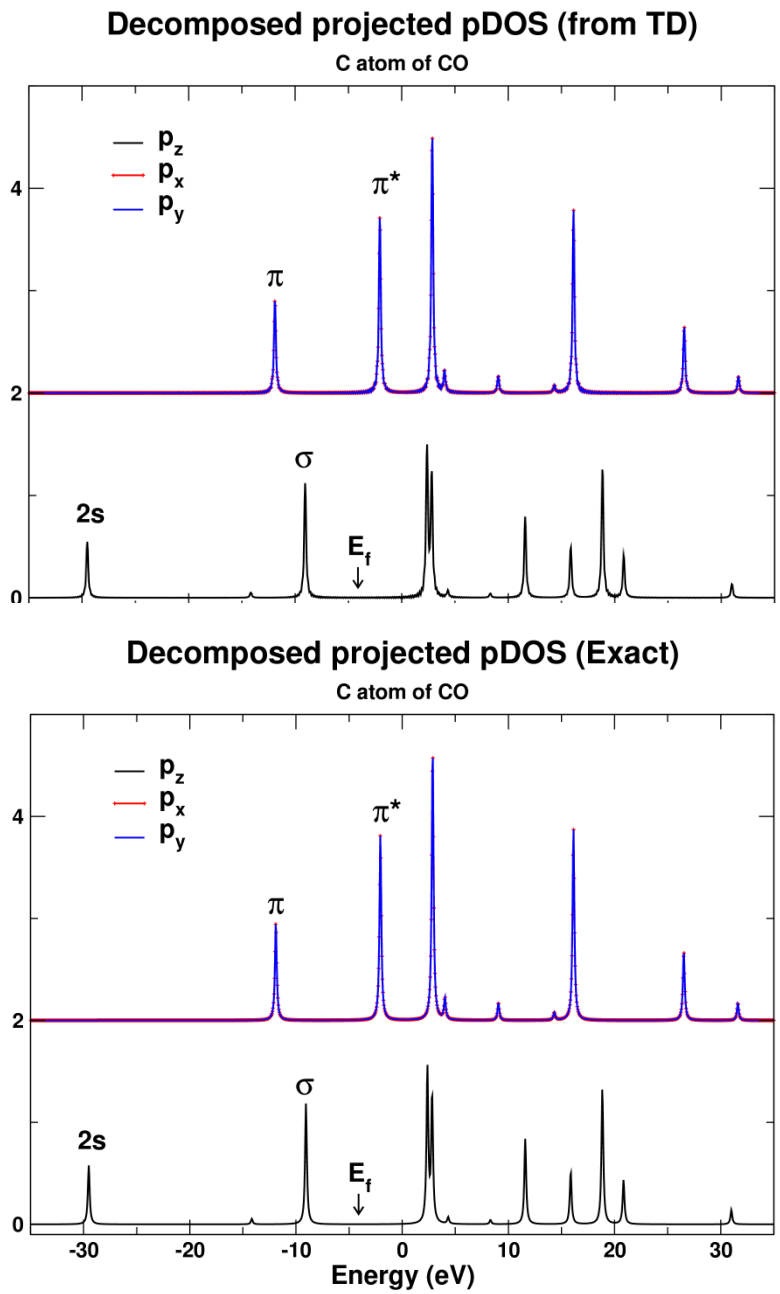
Connection with Fermi golden rule agrees in limit $t \to \infty$

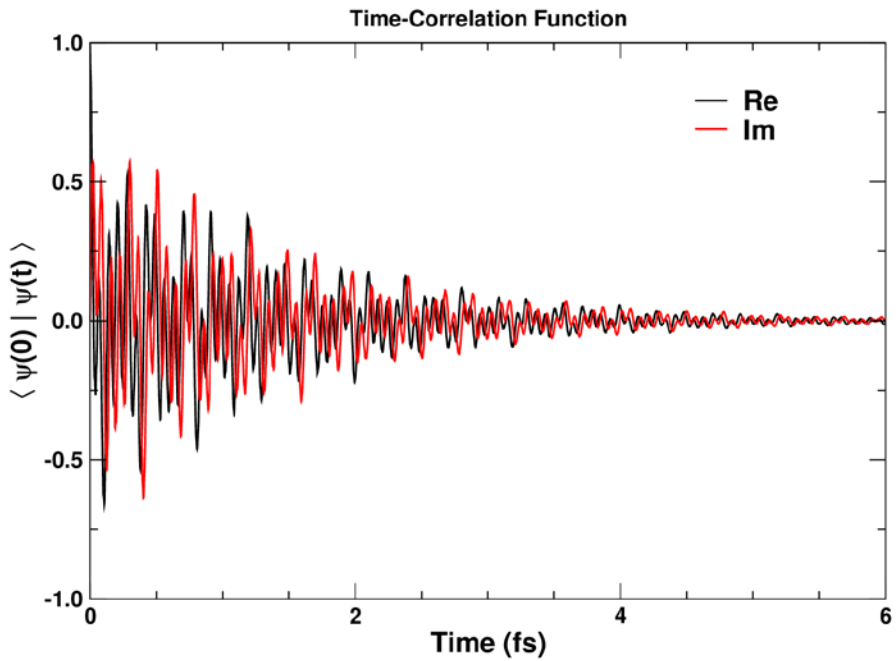
$$G(\omega) = \int_0^\infty dt\, e^{i\omega t} U(t,0) \quad G(E) = [E - H + i\Gamma]^{-1}$$

$$\mu(\omega) = -\frac{1}{\pi} \mathrm{Im} \langle c | d^\dagger G(E) d | c \rangle \theta(E - E_F)$$

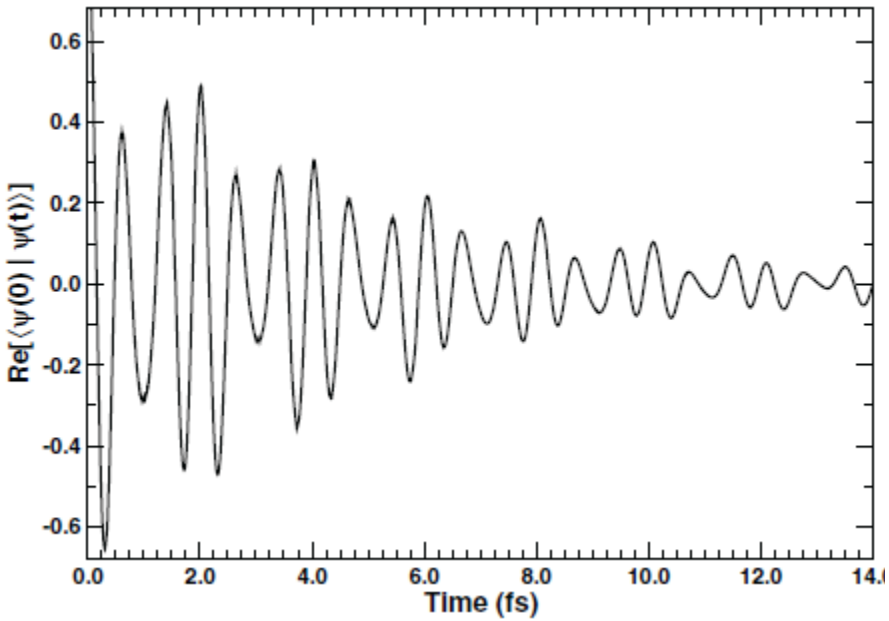
$$\mu(\omega) = \sum_k |\langle c | d | k \rangle|^2 \delta_\Gamma(\omega + \epsilon_c - \epsilon_k) \theta(E - E_F)$$

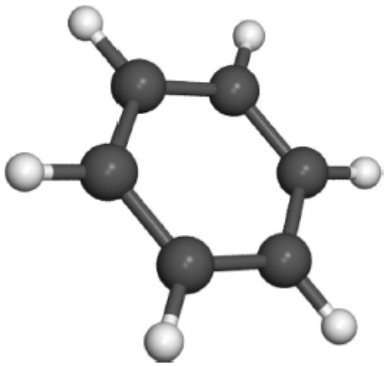
Check:
C K-edge of CO

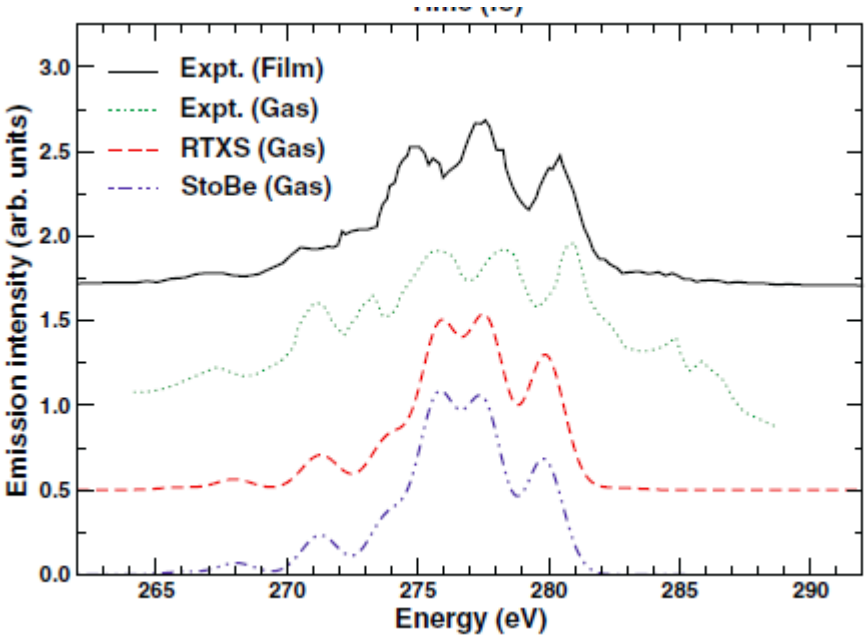




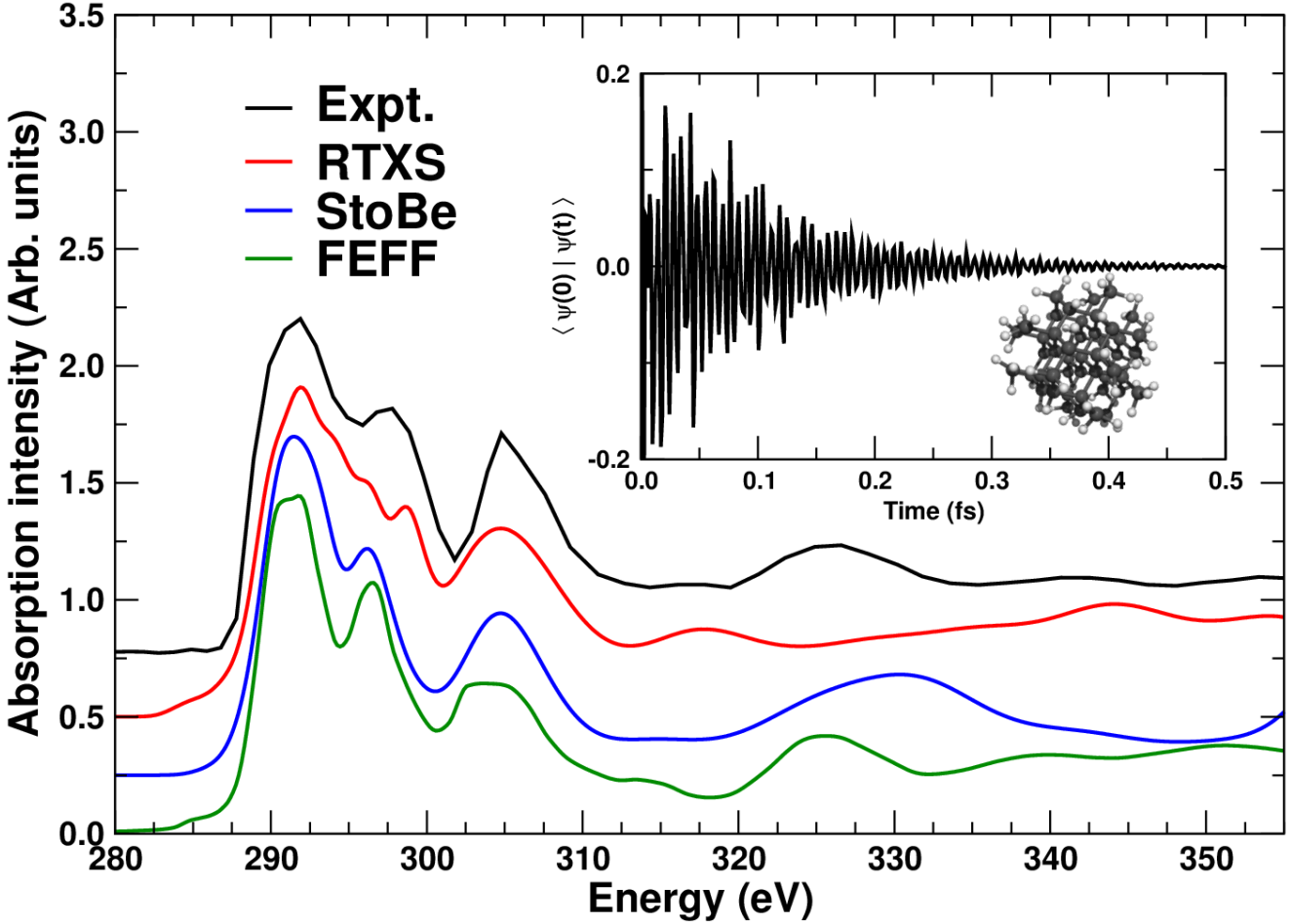
Example: C *K*-edge XES of Benzene







C K-Edge XAS of Diamond ($C_{47}H_{60}$ cluster)



III. Many-body effects: Intrinsic losses

Cumulant expansion for core-hole Green's function*

$$G_c^+(t) = e^{i\epsilon_c t} e^{C(t)} \theta(t)$$

*D. Langreth Phys Rev B 1, 471 (1970)

Particle-hole cumulant for XAS*

PHYSICAL REVIEW B **94**, 035156 (2016)

Particle-hole cumulant approach for inelastic losses in x-ray spectra

J. J. Kas,[1] J. J. Rehr,[1] and J. B. Curtis[2]
[1]*Department of Physics, University of Washington, Seattle, Washington 98195-1560, USA*
[2]*Department of Physics, University of Rochester, Rochester, New York 14927, USA*

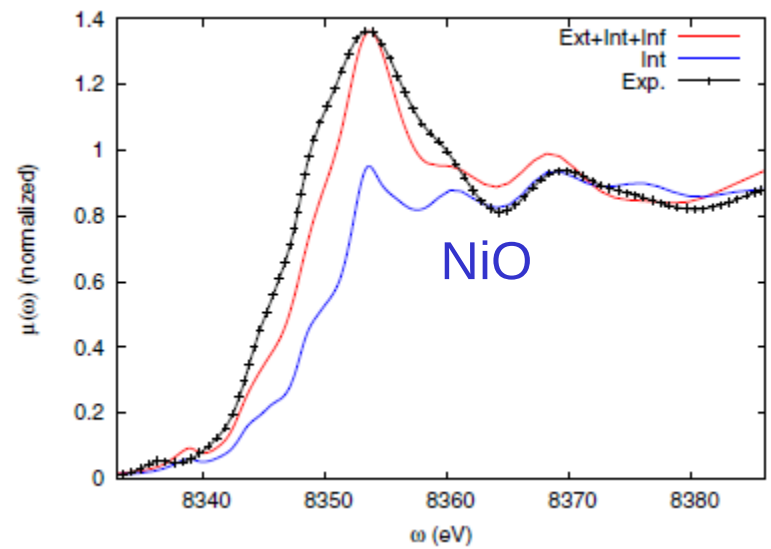
$$\tilde{C}_K(t) = \int d\omega\, \gamma_K(\omega)(e^{i\omega t} - i\omega t - 1)$$

$$\tilde{G}_K(t) = \tilde{G}^0_K(t) e^{\tilde{C}_K(t)}$$

$$\tilde{C}_K(t) = C_c(t) + C_k(t) + C_{ck}$$

All losses in particle-hole spectral function $\boldsymbol{A_K}$

$$\mu(\omega) = \int d\omega'\, \tilde{A}_K(\omega')\mu_K(\omega - \omega')$$



* cf. L. Campbell, L. Hedin, J. J. Rehr, and W. Bardyszewski, Phys. Rev. B **65**, 064107 (2002)

Intrinsic losses: real-time TDDFT cumulant

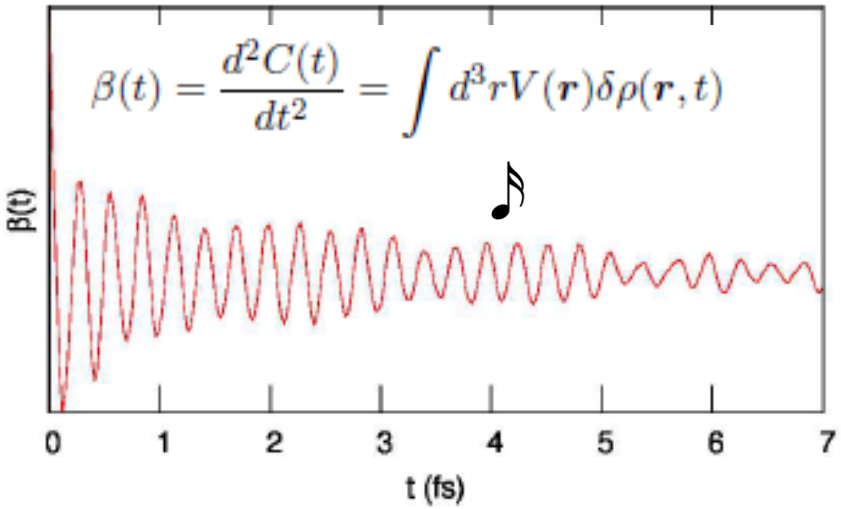
PHYSICAL REVIEW B **91**, 121112(R) (2015)

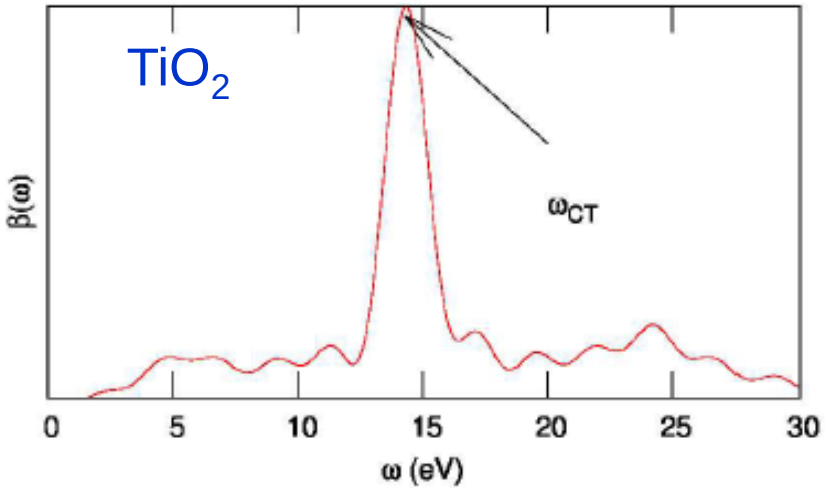
Real-time cumulant approach for charge-transfer satellites in x-ray photoemission spectra

J. J. Kas,[1] F. D. Vila,[1] J. J. Rehr,[1] and S. A. Chambers[2]
[1]*Department of Physics, University of Washington, Seattle, Washington 98195-1560, USA*
[2]*Physical Sciences Division, Pacific Northwest National Laboratory, Richland, Washington 99352, USA*

Langreth cumulant in time-domain*

$$C(t) = \sum_{\boldsymbol{q},\boldsymbol{q}'} V_{\boldsymbol{q}}^* V_{\boldsymbol{q}'} \int d\omega S(\boldsymbol{q},\boldsymbol{q}',\omega) \frac{e^{i\omega t} - i\omega t - 1}{\omega^2} = \int d\omega \beta(\omega) \frac{e^{i\omega t} - i\omega t - 1}{\omega^2}$$



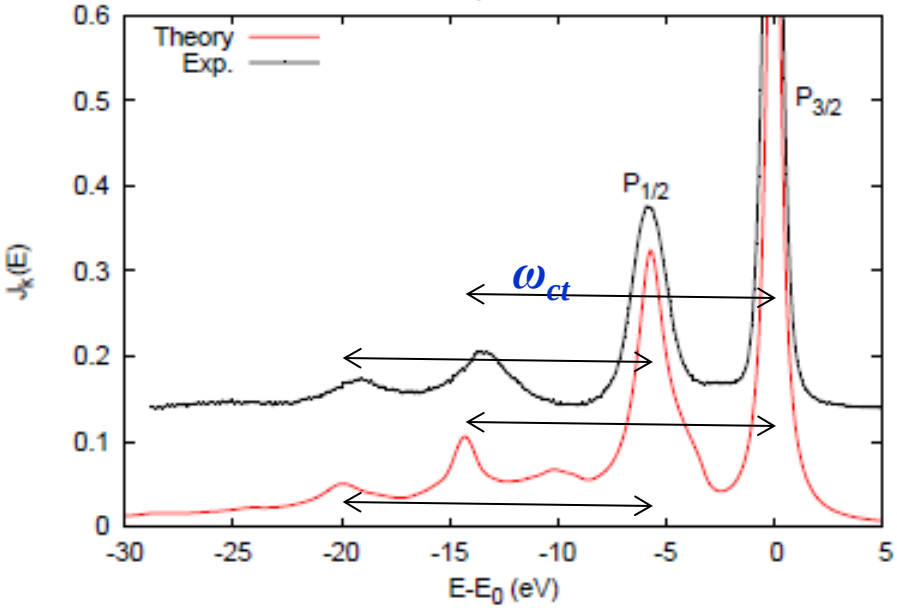


*D. C. Langreth, Phys. Rev. B **1**, 471 (1970)

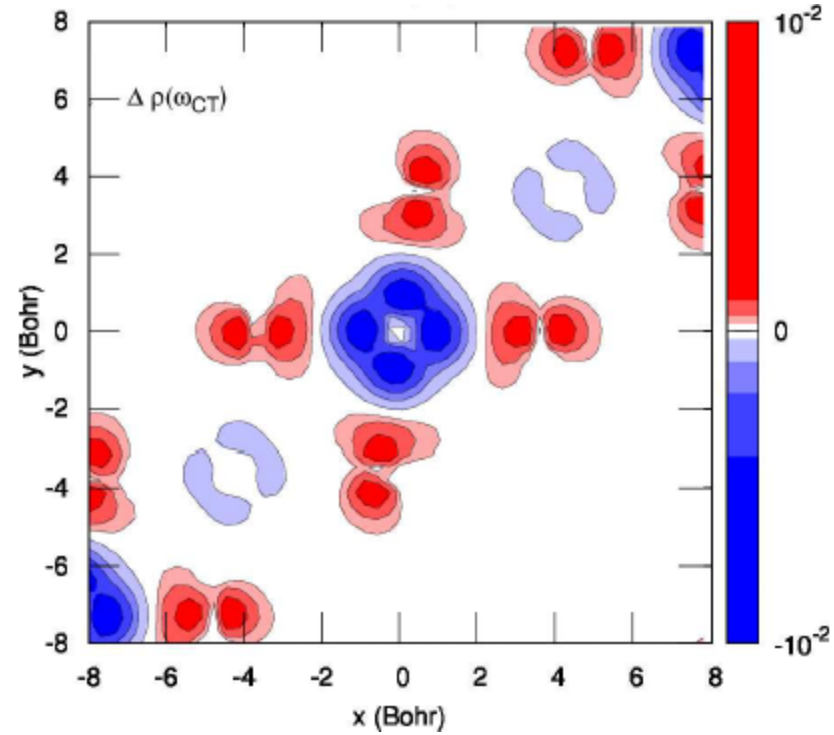
Real-space interpretation: RT-TDDFT cumulant explains intrinsic excitations in TiO_2

RT TDDFT Cumulant

Theory vs XPS



Charge transfer fluctuations



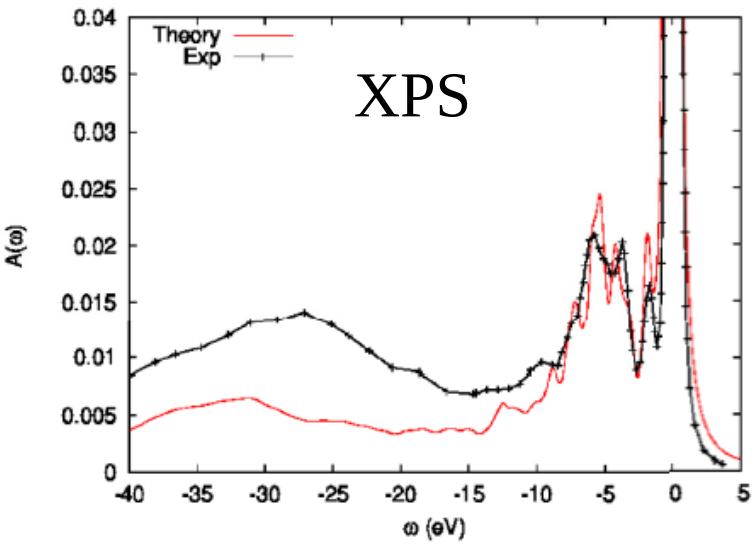
Interpretation: satellites arise from oscillatory charge density fluctuations between ligand and metal at frequency ~ ω_{CT} due to turned-on core-hole

NIST Preprint; submitted to PRB 2016

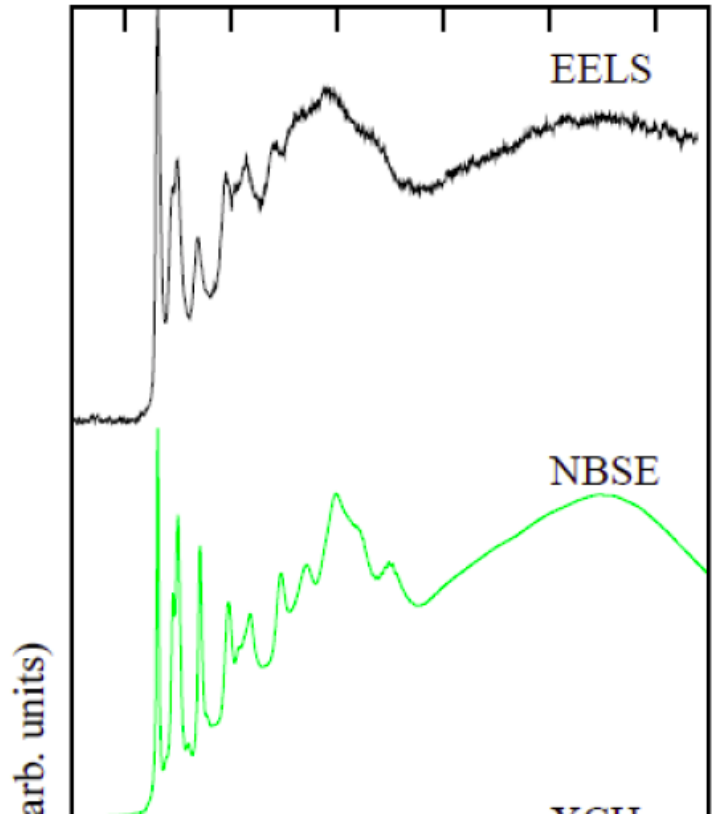
High-resolution valence and core excitation spectra of solid C_{60} via first-principles calculations and experiment

F. Fossard, K. Gilmore, G. Hug, J J. Kas, J J Rehr, E L Shirley and F D Vila

RT-TDDFT cumulant

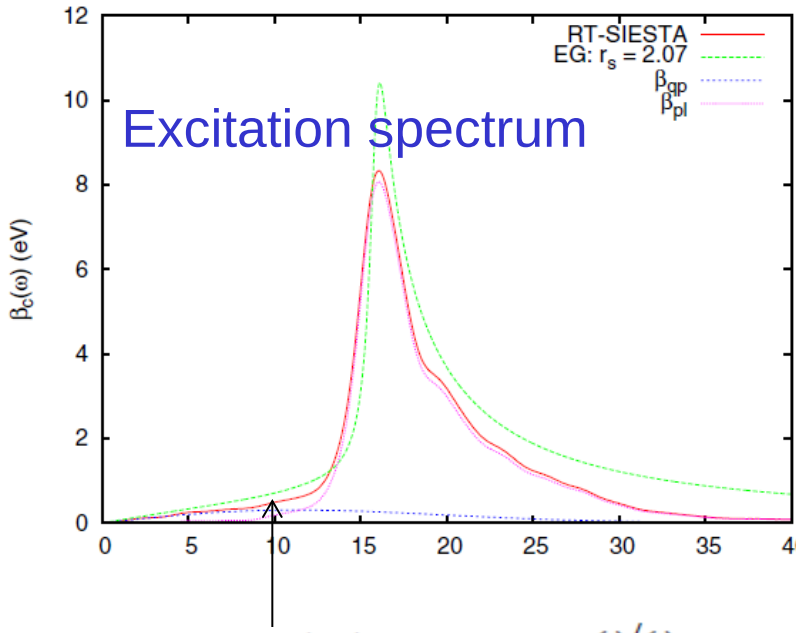


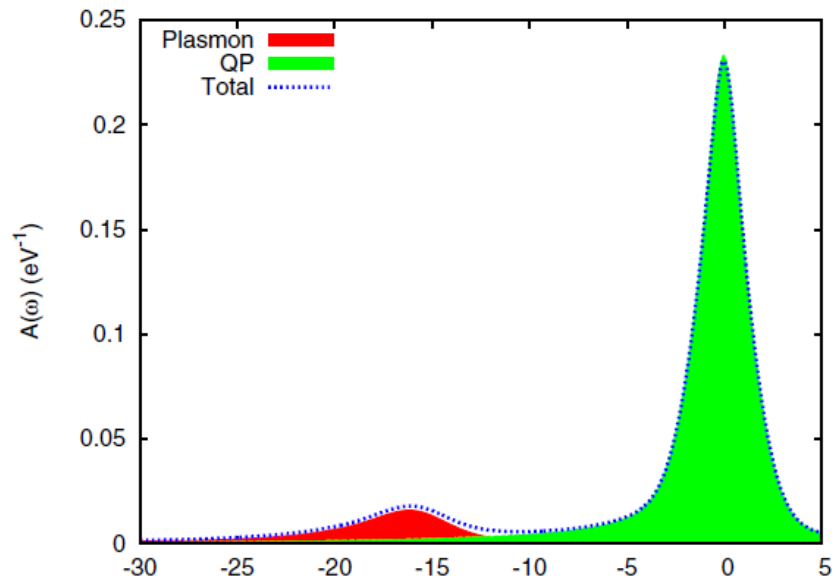
Particle-hole cumulant



X-ray Edge Singularities

Low energy particle-hole excitations in cumulant **explain** edge singularities in XPS and XAS of metals





$$\beta_{ph}(\omega) = \alpha\omega e^{-\omega/\omega_p}$$

$$C_{ph}(t) = -i\alpha\omega_p t - \alpha\ln(1 - i\omega_p t) \qquad A_{ph}(\omega) = e^{-a_{pl}} \frac{e^{-\tilde{\omega}/\omega_p}}{\Gamma(\alpha)} \frac{\omega_p^{-\alpha}}{\tilde{\omega}^{1-\alpha}}$$

Question: **Does the cumulant method work for correlated systems ?**

Hedin's answer * MAYBE

"Calculation similar to core case … but with more complicated fluctuation potentials …

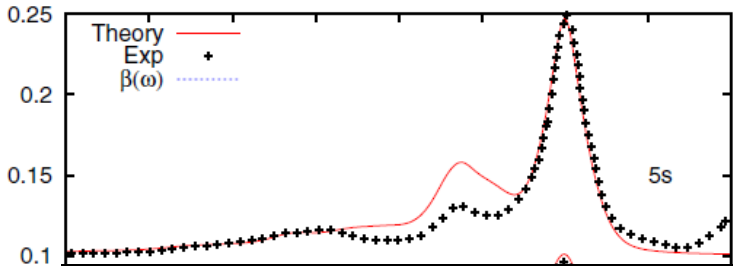
$$V^n \rightarrow -\mathrm{Im}\, \varepsilon^{-1}(\omega_n, q_n)$$

… not question of principle, but of computational work..."

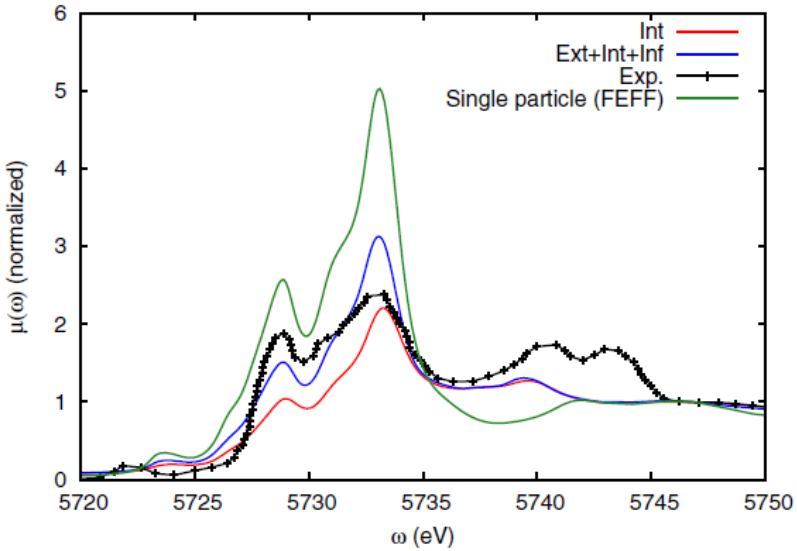
* L. Hedin, J. Phys.: Condens. Matter **11,** R489 (1999)

Particle-hole cumulant for CeO_2

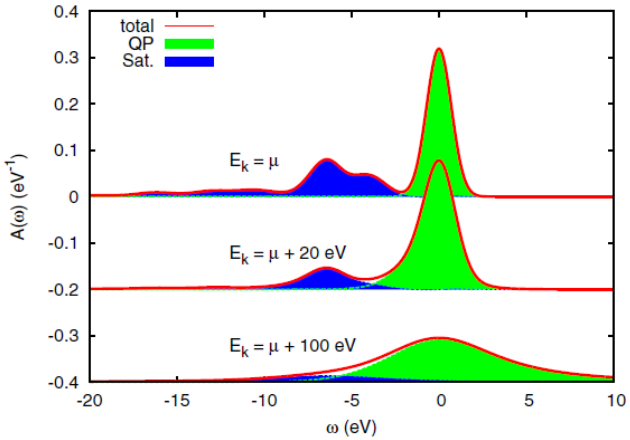
Ce *5s* XPS of CeO_2



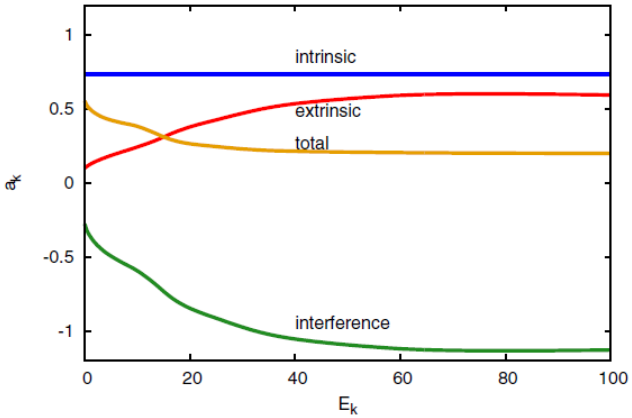
Ce L_3 XAS of CeO_2



Spectral function



Spectral weights



Real-time approach for x-ray Debye-Waller factors

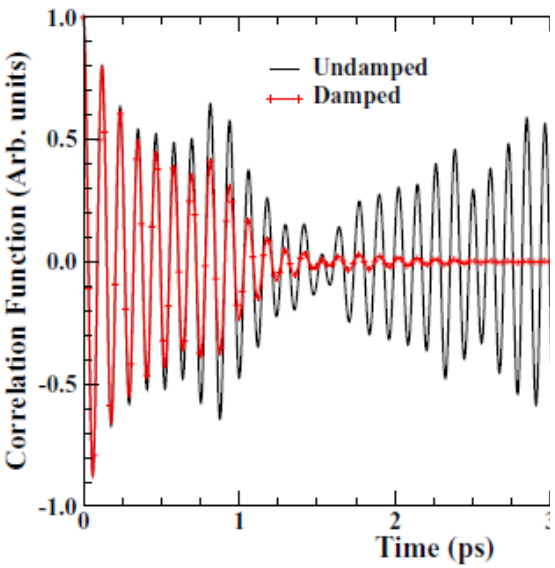
PHYSICAL REVIEW B **85**, 024303 (2012)

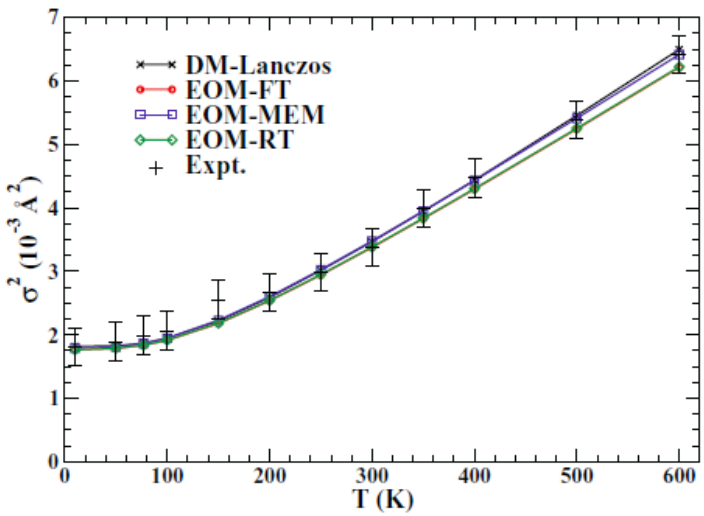
X-ray absorption Debye-Waller factors from *ab initio* molecular dynamics

F. D. Vila, V. E. Lindahl, and J. J. Rehr
Department of Physics, University of Washington, Seattle, Washington 98195, USA
(Received 30 August 2011; revised manuscript received 11 January 2012; published 25 January 2012)

An *ab initio* equation of motion method is introduced to calculate the temperature-dependent mean-squa vibrational amplitudes σ^2 which appear in the Debye-Waller factors in x-ray absorption, x-ray scattering, a related spectra. The approach avoids explicit calculations of phonon modes, and is based instead on calculatio of the displacement-displacement time correlation function from *ab initio* density functional theory molecula dynamics simulations. The method also yields the vibrational density of states and thermal quantities such as the lattice free energy. Illustrations of the method are presented for a number of systems and compared with other methods and experiment.

$$\sigma_R^2(T) = \frac{\hbar}{\mu_R \pi} \int_0^{t_{\max}} dt \langle Q_R(t) | Q_R(0) \rangle \times \ln\left[\left(2\sinh\frac{\pi t}{\beta\hbar}\right)^{-1}\right] e^{-\varepsilon t^2}$$





Conclusions

Efficient RT-TDDFT approach for frequency dependent nonlinear optical response –

Accuracy comparable to frequency-domain methods for small systems; also applicable to large systems

Similar real-time approaches can be applied to dynamic structure, Debye-Waller factors , etc.

Acknowledgments:

Supported by DOE BSE DE-FG02-97ER45623

Thanks to

J.J. Kas	**L. Reining**	**G. Bertsch**
J. Vinson	K. Gilmore	L. Campbell
T. Fujikawa	F. Vila	E. Shirley
S. Story	S. Biermann	M Guzzo
M. Verstraete et al.	J. Sky Zhou	C. Draxl

& especially the ETSF

Conclusions

Particle-hole cumulant theory yields reasonable approximation for inelastic losses in XPS & XAS

$$\mu(\omega) = \int d\omega' \, \tilde{A}_K(\omega') \mu_K(\omega - \omega')$$

All losses (intrinsic, extrinsic and interference) in spectral function $\boldsymbol{A_K}(\omega)$ – *can be added ex post facto*

Interference terms explain mysteries in amplitudes and energy dependence: adiabatic- sudden transition

Theory also applicable to some *d*- and *f*-systems.

Many-body amplitudes $S_0^2(\omega)$ in XAS

- Many-body XAS ≈ Convolution

$$\mu(\omega) = \int_0^\infty d\omega' \tilde{A}(\omega,\omega')\mu_{qp}(\omega-\omega')$$

$$\equiv \langle \mu_{qp}(\omega)\rangle \approx \mu_{qp}(\omega)\, S_0^2(\omega)$$

- Explains crossover: **adiabatic** $S_0^2(\omega) = 1$

  to **sudden** transition $S_0^2(\omega) \approx 0.9$

$$|g_q|^2 = |g_q^{ext}|^2 + |g_q^{intrin}|^2 - 2\, g_q^{ext}\, g_q^{intrin}$$

Interference reduces loss!

Dynamic core-hole screening algorithm

PHYSICAL REVIEW B 73, 075402 (2006)

Dynamical core-hole screening in the x-ray absorption spectra of graphite, C_{60}, and carbon nanotubes: A first-principles electronic structure study

O. Wessely and O. Eriksson
Department of Physics, Uppsala University, Box 530, SE-75121, Uppsala, Sweden

M. I. Katsnelson
Institute for Molecules and Materials, Radboud University Nijmegen, NL-6525 ED Nijmegen, The Netherlands

(Received 14 September 2005; revised manuscript received 23 November 2005; published 1 February 2006)

alá Nozieres & De Dominicis

$$I(\omega) \sim \mathrm{Re} \int_0^\infty dt \int_\mu^\infty \frac{d\epsilon}{\pi} e^{it(\omega-\epsilon+\epsilon_c)+\Delta(-t)} \times \sum_{ij} t_i [\mathrm{Im}\, \mathbf{g}^R(\epsilon)\bar{\varphi}(\epsilon,t)]_{ij} t_j^* , \qquad (8)$$

$$\Delta(t) = i\, \mathrm{Tr}\, \mathbf{V} \int_0^{-t} dt_1 \int_{-\infty}^{\mu} \frac{d\epsilon}{\pi} \mathrm{Im}\, \mathbf{g}^R(\epsilon)\bar{\varphi}(\epsilon,t_1), \qquad (9)$$

where $\bar{\varphi}(\epsilon,t)$ can be found from the matrix integral equation

$$\bar{\varphi}(\epsilon,t) = \mathbf{1} + \mathbf{V} \int_\mu^\infty \frac{d\epsilon_1}{\pi} \mathrm{Im}\, \mathbf{g}^R(\epsilon_1) \mathbf{L}^+(\epsilon,\epsilon_1,t)\bar{\varphi}(\epsilon_1,t),$$

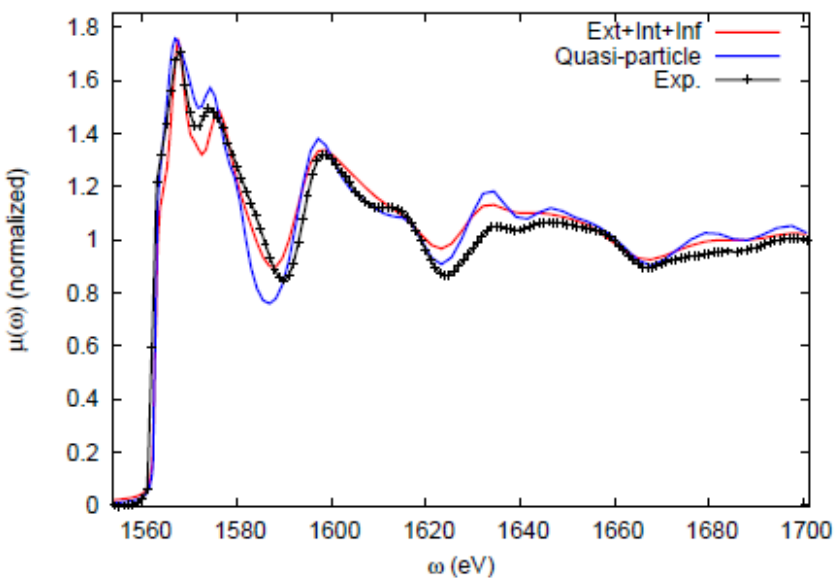
$$\mathbf{L}^+(\epsilon,\epsilon_1,t) = \frac{\mathbf{I}^*(\epsilon_1,t) - e^{i(\epsilon-\epsilon_1)t}\mathbf{I}^*(\epsilon,t)}{\epsilon_1 - \epsilon},$$

$$\mathbf{I}(\epsilon,t) = \mathbf{1} + \mathbf{V} \int_{-\infty}^{\infty} \frac{d\epsilon_1}{\pi} \frac{e^{i(\epsilon-\epsilon_1)t} - 1}{\epsilon - \epsilon_1} \mathrm{Im}\, \mathbf{G}^R(\epsilon_1). \qquad (10)$$

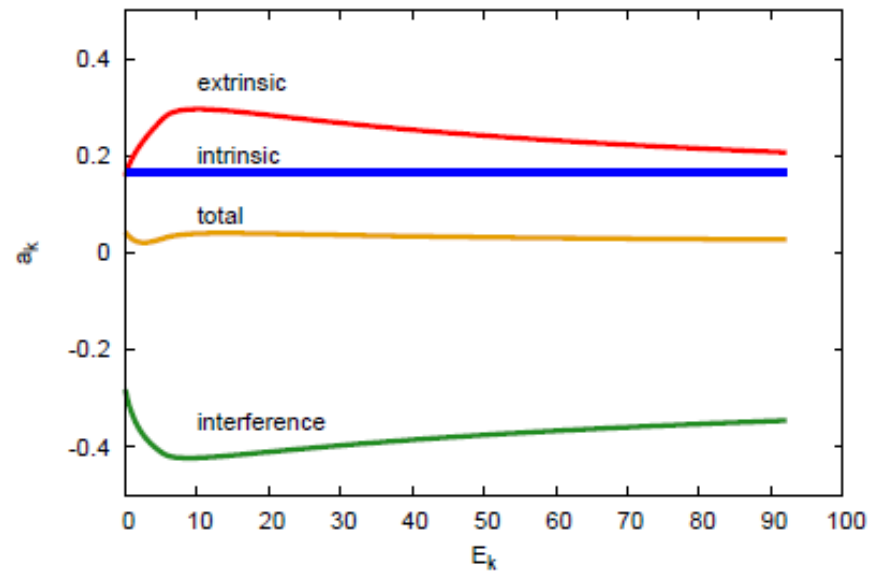
Also: Grebennikov, Babanov and Sololov, Phys. Stat. Sol. **79**, 423 (1977)
and Privalov, Gel'mukhanov & Agren: Phys Rev. B **64,** 165115 (2001)

Extrinsic losses and Interference

XAS of Al



Satellite strengths



Particle-hole cumulant explains **cancellation** of extrinsic and intrinsic losses at threshold and

crossover: **adiabatic**

to sudden approximation

Extension to: *Extrinsic and Intrinsic losses*

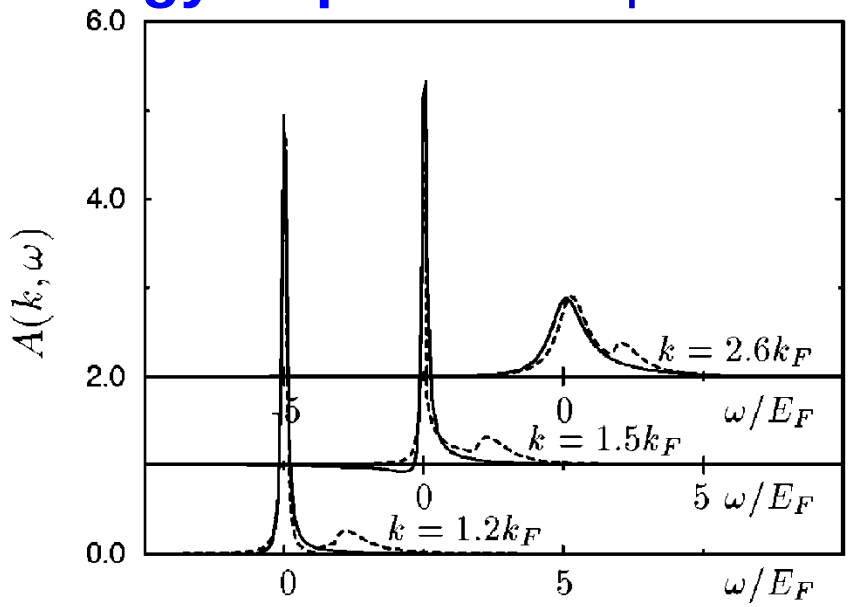
Quasi-boson Model

PHYSICAL REVIEW B, VOLUME 65, 064107

Interference between extrinsic and intrinsic losses in x-ray absorption fine structure

L. Campbell,[1] L. Hedin,[2] J. J. Rehr,[1] and W. Bardyszewski[3]
[1]*Department of Physics, University of Washington, Seattle, Washington 98195-1560*
[2]*Department of Physics, Lund University, Lund, S22362 Sweden*
and MPI-FKF, Stuttgart, D70569 Germany
[3]*Department of Physics, Institute of Theoretical Physics, 00-681 Warsaw, Poland*

Energy Dependent Spectral Function $A(k,\omega)$



Question: How to *extend* theory to real-time approach?

Interference: Quasi-Boson Approach*

Excitations - plasmons, electron-hole pairs ... are **bosons**

Many-body Model: $|e^-, h, \text{bosons}\rangle$

- Excitations: $H_v = \Sigma_n \omega_n a_n^\dagger a_n$
- Electrons: $h' = \Sigma_k \epsilon_k c_k^\dagger c_k$
- e-boson coupling $V_{pv} = \Sigma_{nkk'} \left[V_{kk'}^n a_n^\dagger + (V_{kk'}^n)^* a_n \right] c_k^\dagger c_{k'}$
- Core-hole-boson coupling: $V_{vc} = -\Sigma_n V_{bb}^n \left(a_n^\dagger + a_n \right)$

"GW++" **Same ingredients as GW self-energy**

$V^n \rightarrow -\mathrm{Im}\, \varepsilon^{-1}(\omega_n, q_n)$ fluctuation potentials

*W. Bardyszewski and L. Hedin, Physica Scripta **32**, 439 (1985)

*Many-pole Self-energy Algorithm**

Plasmon-pole model → many-pole model

$-\mathrm{Im}\,\varepsilon^{-1}(\omega)$ *Many-pole* Dielectric Function

→ $\sim \Sigma_i g_i \delta(\omega - \omega_i)$

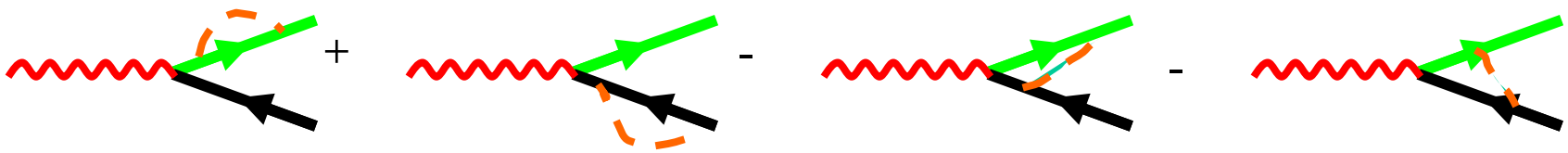
→ *Many-pole* GW self-energy $\Sigma(E)$

* J. Kas et al. PRB **76**, 195116(2008)

Effective GW++ Green's Function $g_{eff}(\omega)$

L. Campbell, L. Hedin, J. J. Rehr, and W. Bardyszewski, Phys. Rev. B **65**, 064107 (2002)

$$g_{eff}(\omega) = e^{-a}\left[g'(\omega) + \Sigma_n \left(\frac{V^n_{bb}}{\omega_n}\right)^2 g'(\omega-\omega_n) - 2\Sigma_n \frac{V^n_{bb}}{\omega_n} g'(\omega-\omega_n) V^n g'(\omega)\right]$$



Extrinsic + Intrinsic - 2 x Interference

Damped ***qp*** Green's function $g'(\omega) \equiv [\omega - h' - \Sigma(\omega) + i\gamma]^{-1}$

Spectral function: $A(\omega) = -(1/\pi)\ \mathrm{Im}\ g_{eff}(\omega)$

That's all folks