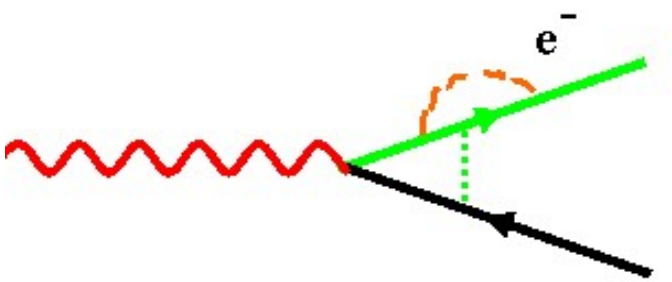
Perfecting the Theory of XAFS

J. J. Rehr

University of Washington and SLAC



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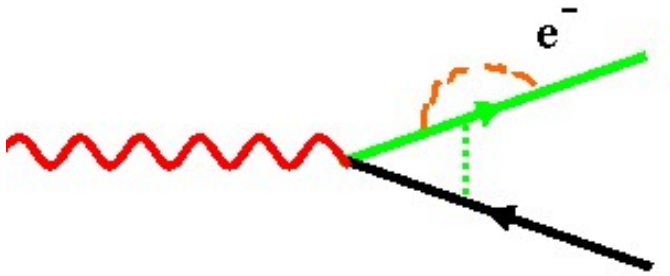
Perfecting the Theory of XAFS

- **TALK -**



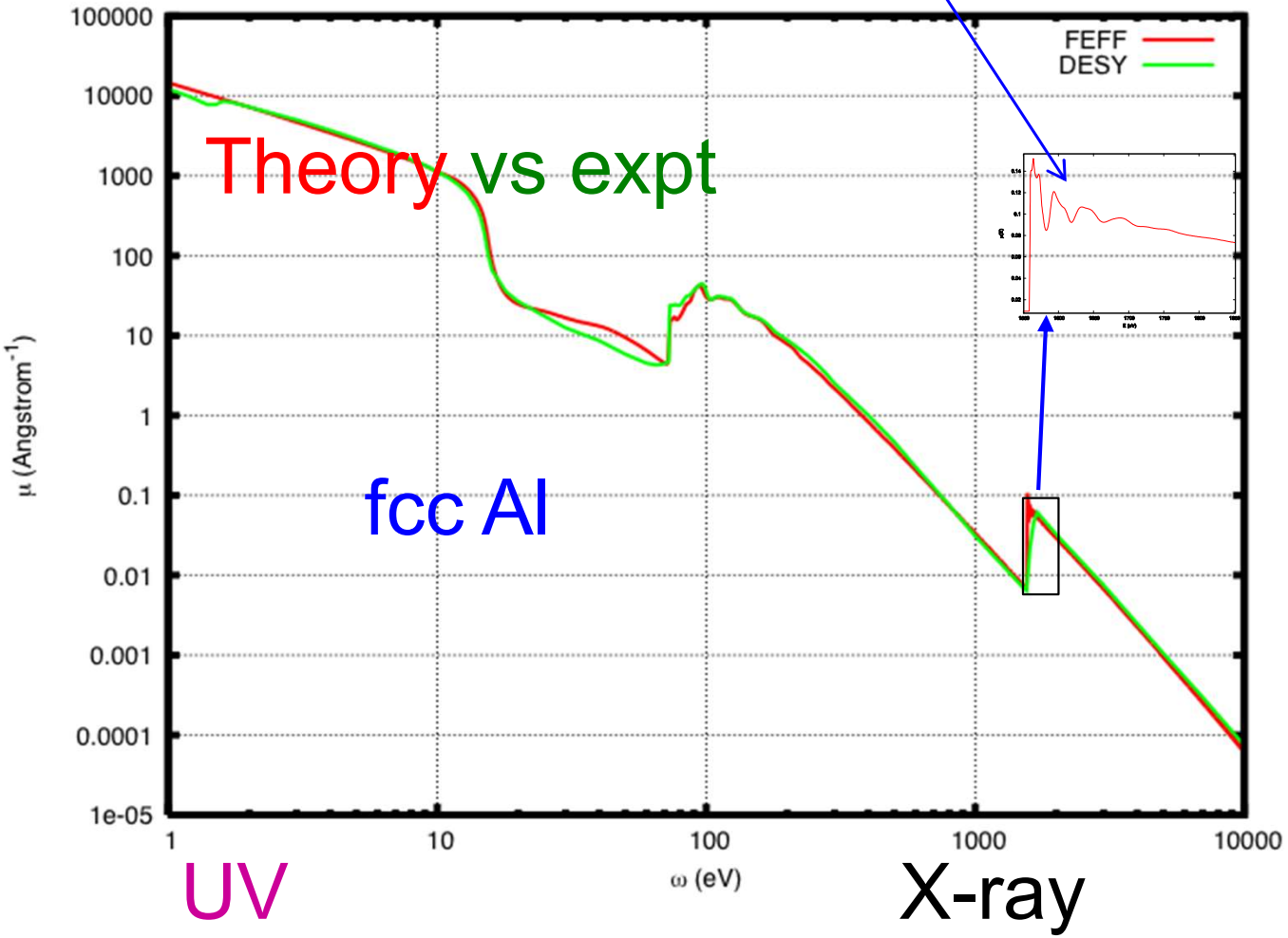
I. Challenges in XAFS Theory

II. Theoretical advances over 50 years

III. Beyond XAFS - XANES, Multiplets, BSE & Corvus Workflows

I. Challenge of XAFS theory

X-ray Absorption Fine Structure



UV

X-ray

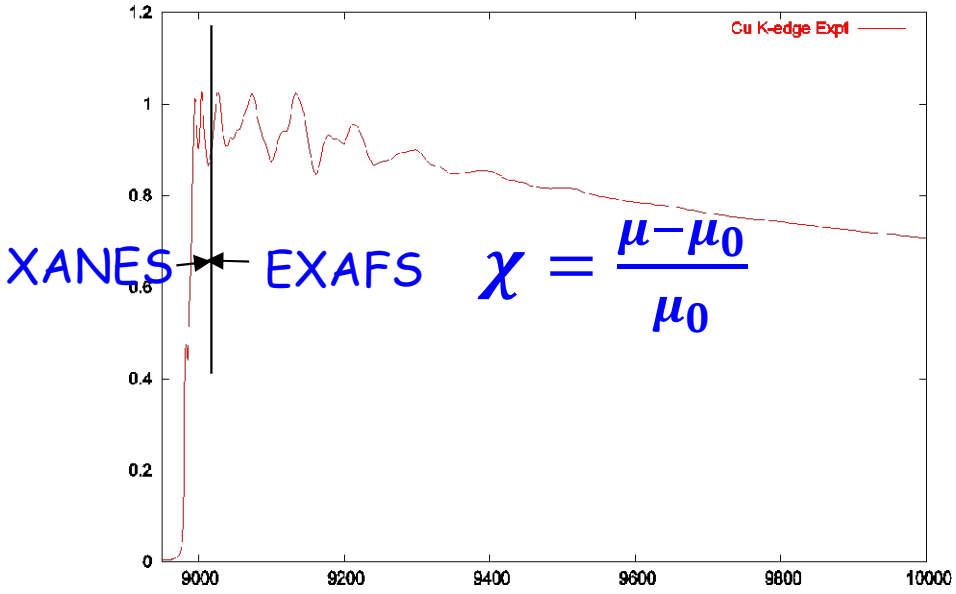
Photon energy (eV)

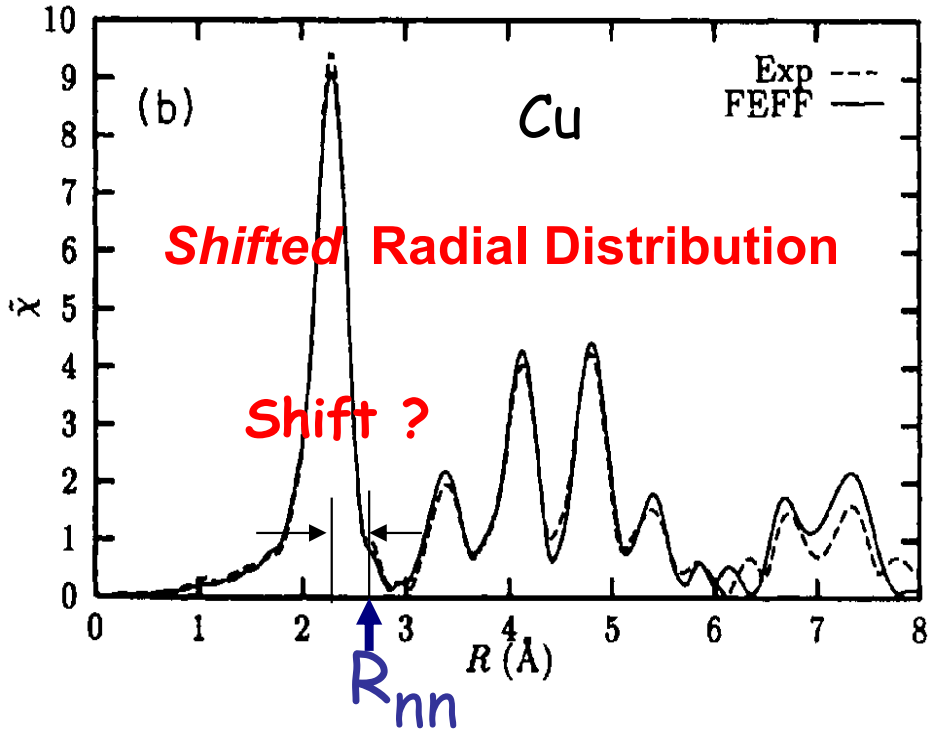
$$\chi = \frac{\mu - \mu_0}{\mu_0}$$

Small correction to atomic XAS - need high accuracy & very large energy range

Historical Interpretation ~ 50 yrs ago*

EXAFS → FT = Local structure





*Stern Sayers Lytle, UW
PRL **27**, 1204 (1971)

→ X-ray Microscope!

XAFS Eq. $\chi(k) = -kf(k)\sum_j [N_j \exp(-\gamma r_j)/r_j^2] \exp(-\sigma_j^2 k^2/2) \sin[2kr_j + 2\eta(k)]$

Many parameters - need to calibrate with "EXAFS standard"

Question: Can one calculate XAFS parameters?

Maybe or maybe not ?

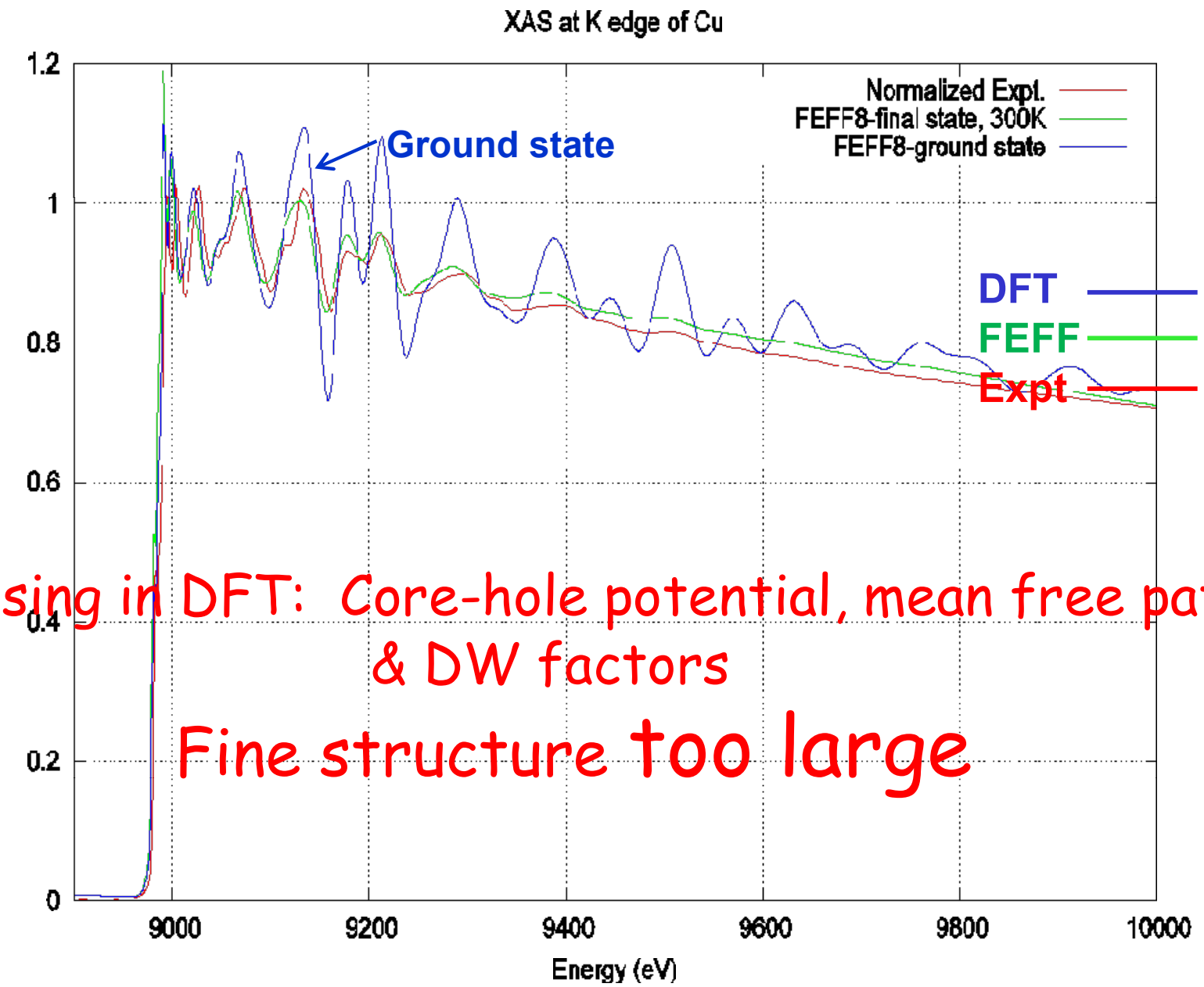
”I always thought it was easier to

measure XAS than to calculate it.”

Hans Bethe

ca 1980

Standard electronic structure theory *FAILS*



Missing in DFT: Core-hole potential, mean free path & DW factors

Fine structure too large

II. Theoretical Advances

Breakthrough: Spherical wave scattering theory
FIXES Plane Wave Approximation

PHYSICAL REVIEW B VOLUME 34, NUMBER 6 15 SEPTEMBER 1986

New high-energy approximation for x-ray-absorption near-edge structure

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C. R. Natoli
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Asymptotic high energy approximation:
effective spherical wave scattering amplitudes f_{eff}

R_f for XAFS

Revised EXAFS Equation: Stern, Sayers, Lytle

$$\chi(k) = S_0^2 \sum_R \frac{|f_{\text{eff}}(k)|}{kR^2} \sin(2kR + \Phi_k) e^{-2R/\lambda_k} e^{-2\sigma^2 k^2}$$

Curved wave scattering theory $k \sim 25$ Å^{-1} $l \sim 25$ f_{eff} Φ_k

Mean free paths $\lambda_k \sim 5 - 20$ Å **and self energy** $\Sigma(E)$

Vibrational damping $e^{-2\sigma^2 k^2}$

Multi-electron excitations $S_0^2 \sim 0.8$

Relativistic scattering potentials $V(r)$

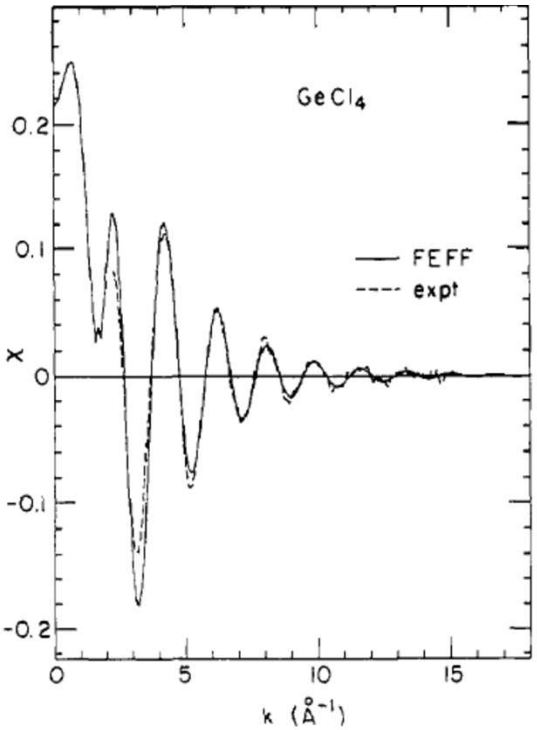
No codes in 1970's with all features !

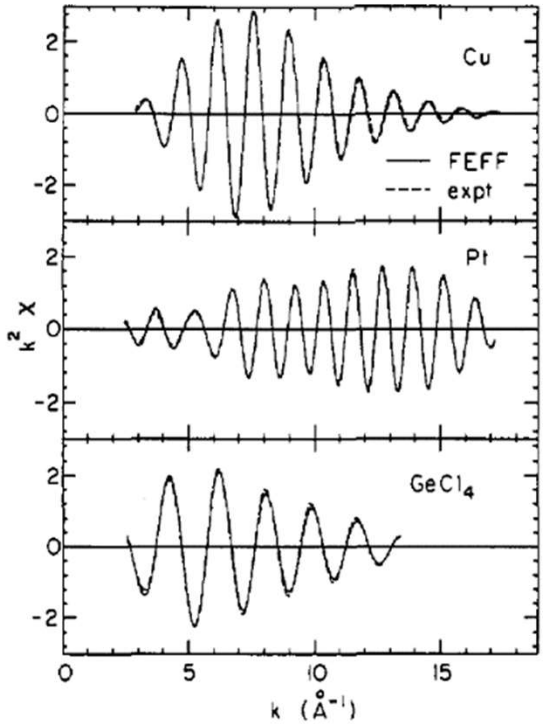
Three key advances

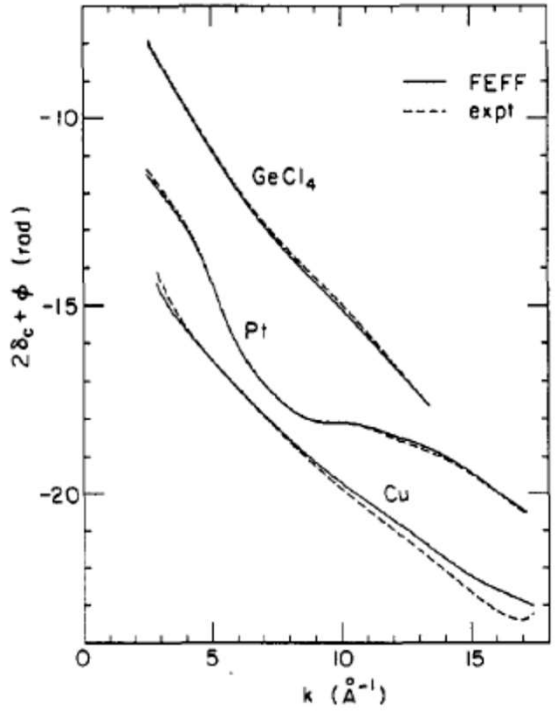
Curved wave single-scattering amplitudes f_{eff} & phases φ

Hedin-Lundqvist Self-energy $\Sigma(E)$

Relativistic Dirac-Fock scattering potentials $V(r)$







Approximations: Overlapped atom potentials, const S_0^2

Result: Theoretical XAFS Standards

Theoretical X-ray Absorption Fine Structure Standards

JACS **113**, 5136 **(1991)**

J. J. Rehr,*,† J. Mustre de Leon,†,‡ S. I. Zabinsky,† and R. C. Albers§

Contribution from the Department of Physics, FM-15, University of Washington, Seattle, Washington 98195, and Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545. *Received November 13, 1990*

Abstract: Theoretical X-ray absorption fine structure (XAFS) standards are developed for arbitrary pairs of atoms throughout the periodic table ($Z \leq 94$). These standard XAFS spectra are obtained from *ab initio* single-scattering XAFS calculations, using an automated code, FEFF, which takes into account the most important features in current theories: (i) an exact treatment of curved-wave effects; (ii) approximate molecular potentials derived from relativistic atoms, (iii) a complex, energy-dependent self-energy; (iv) a well defined energy reference. FEFF also yields tables of XAFS phases and amplitudes as well as mean-free paths. Sample results are presented and compared with experimental results and with earlier work. We find that these theoretical standards are competitive with experimental standards, permitting XAFS analysis at lower wavenumbers and yielding distance determinations typically better than 0.02 Å and coordination numbers typically better than 20%. These standards also provide theoretical tests of chemical transferability in XAFS.

$$\chi_l^{(1)}(E) = -N\,A(E)\frac{|f_{\mathrm{eff}}(\pi,k,R)|}{kR^2}\sin(2kR + 2\delta_c + \Phi)e^{-2R/\lambda}e^{-2\sigma^2k^2}$$

FEFF3 Accurate single-scattering standards

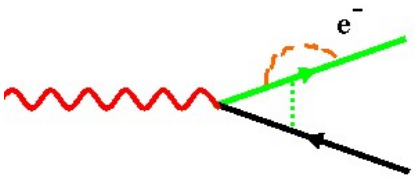
Challenge: Multiple-scattering in XAFS

Golden rule for XAS via Wave functions

$$\mu(E) \sim \Sigma_f |\langle i|\hat{\epsilon}\cdot\mathbf{r}|f\rangle|^2 \delta(E - E_f)$$

Golden rule via Green's functions: Real space MS

$$\mathbf{G} = 1/(E - h' - \Sigma)$$



$$\mu(E) \sim -\tfrac{1}{\pi}\mathrm{Im}\,\langle \mathrm{i}|\,\hat{\epsilon}\cdot\mathbf{r}'\,\mathrm{G}(\mathbf{r}',\mathbf{r},\mathrm{E})\,\hat{\epsilon}\cdot\mathbf{r}\,|\mathrm{i}\rangle$$

h' Final-state Hamiltonian with core-hole

Σ Self-energy including mean-free-path

Multiple-scattering path expansion

$$\mu(E) \sim -\frac{1}{\pi}\mathrm{Im}\,\langle \mathrm{i}|\,\hat{\epsilon}\cdot\mathbf{r}'\,\mathrm{G}(\mathbf{r}',\mathbf{r},\mathrm{E})\,\hat{\epsilon}\cdot\mathbf{r}\,|\mathrm{i}\rangle$$

$$G = G^c + \sum_{i\neq 0} G^c t_i G^c + \sum_{i\neq j \;\; i,j\neq 0} G^c t_i G^0 t_j G^c + \cdots$$

$$\equiv \sum_{\Gamma} G^{\Gamma}.$$

Computational bottleneck *N~100, lmax~20*

Matrix products $Nlmax^2 \times Nlmax^2 \sim 10^4 \times 10^4$

Exact form useful only for low order 4-leg MS paths

Breakthrough: RA Separable Green's function

PHYSICAL REVIEW B VOLUME 41, NUMBER 12 15 APRIL 1990-II

Scattering-matrix formulation of curved-wave multiple-scattering theory: Application to x-ray-absorption fine structure

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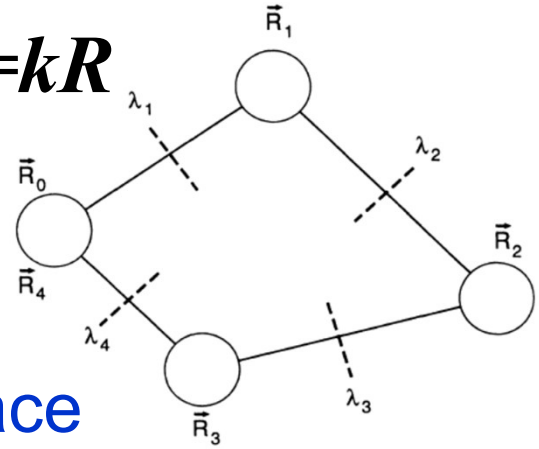
(Received 13 January 1989; revised manuscript received 4 January 1990)

$$G_{L,L'}(\rho)=\frac{e^{i\rho}}{\rho}\sum_{\lambda}\tilde{\Gamma}^{L}_{\lambda}(\rho)\Gamma^{L'}_{\lambda}(\rho)$$

$\rho=kR$



Fast, accurate GF and high-order real-space multiple-scattering *path expansion*

Cut diagrams

Breakthrough: Full multiple scattering - XANES

PHYSICAL REVIEW B VOLUME 58, NUMBER 12 15 SEPTEMBER 1998-II

Real-space multiple-scattering calculation and interpretation of x-ray-absorption near-edge structure

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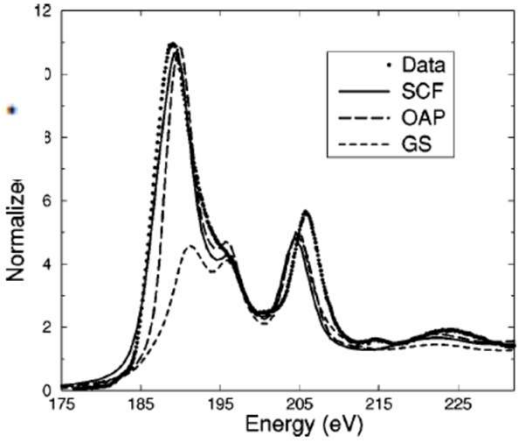
Full multiple scattering by matrix inversion + SCF potentials

$$G^{SC} = G^0 t G^0 + G^0 t G^0 t G^0 + \cdots$$
$$= G^0 (1 - t G^0)^{-1}$$



Implementation – FEFF6

PHYSICAL REVIEW B VOLUME 52, NUMBER 4 15 JULY 1995-II

Multiple-scattering calculations of x-ray-absorption spectra

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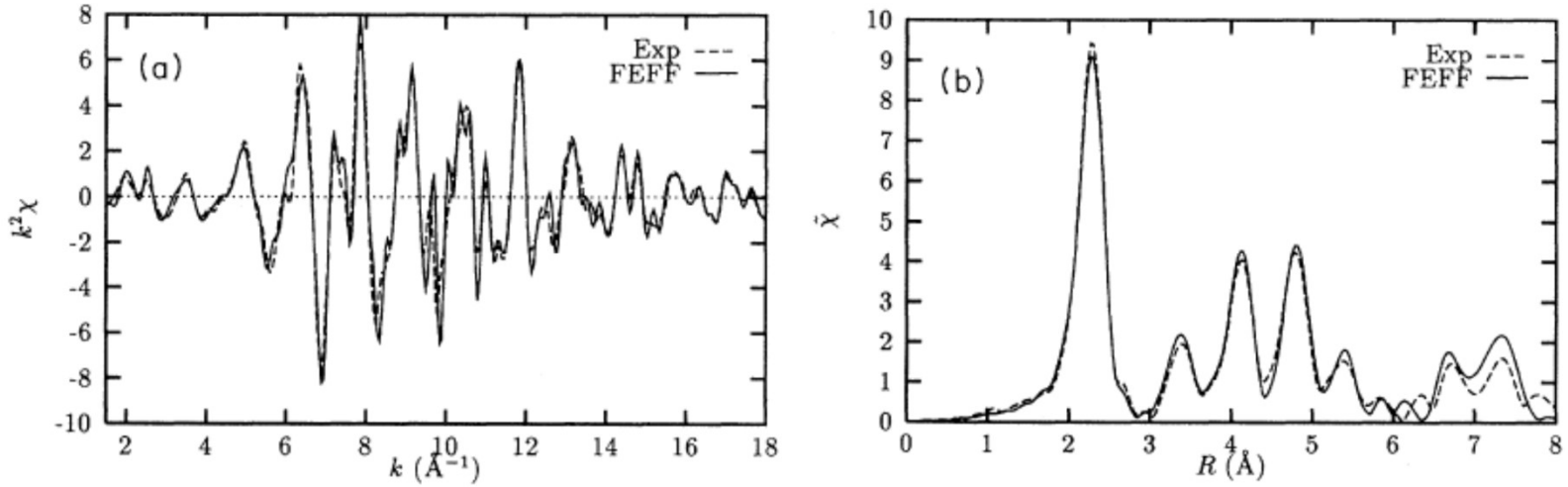
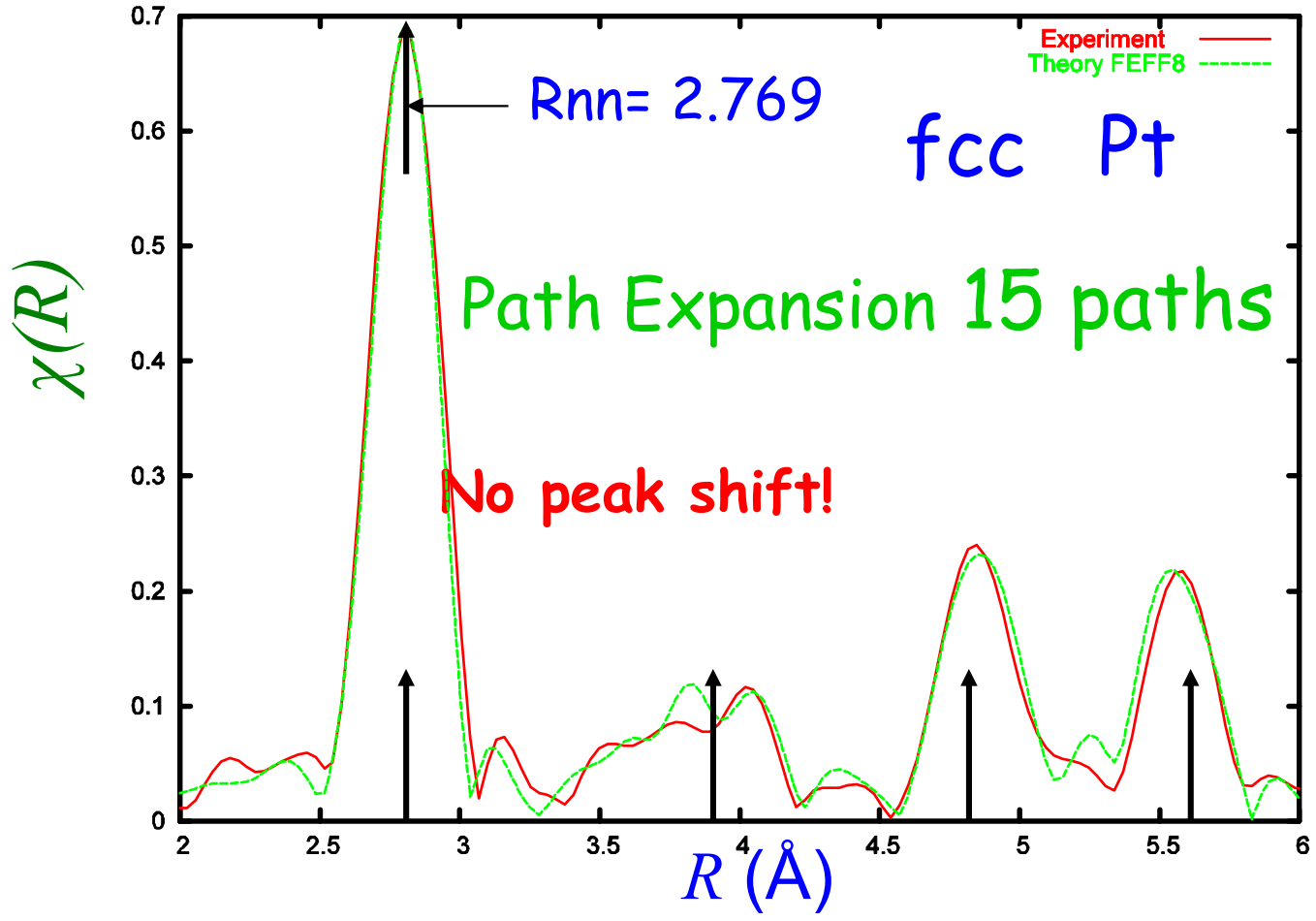


Efficient MS path filters, algorithms and coding

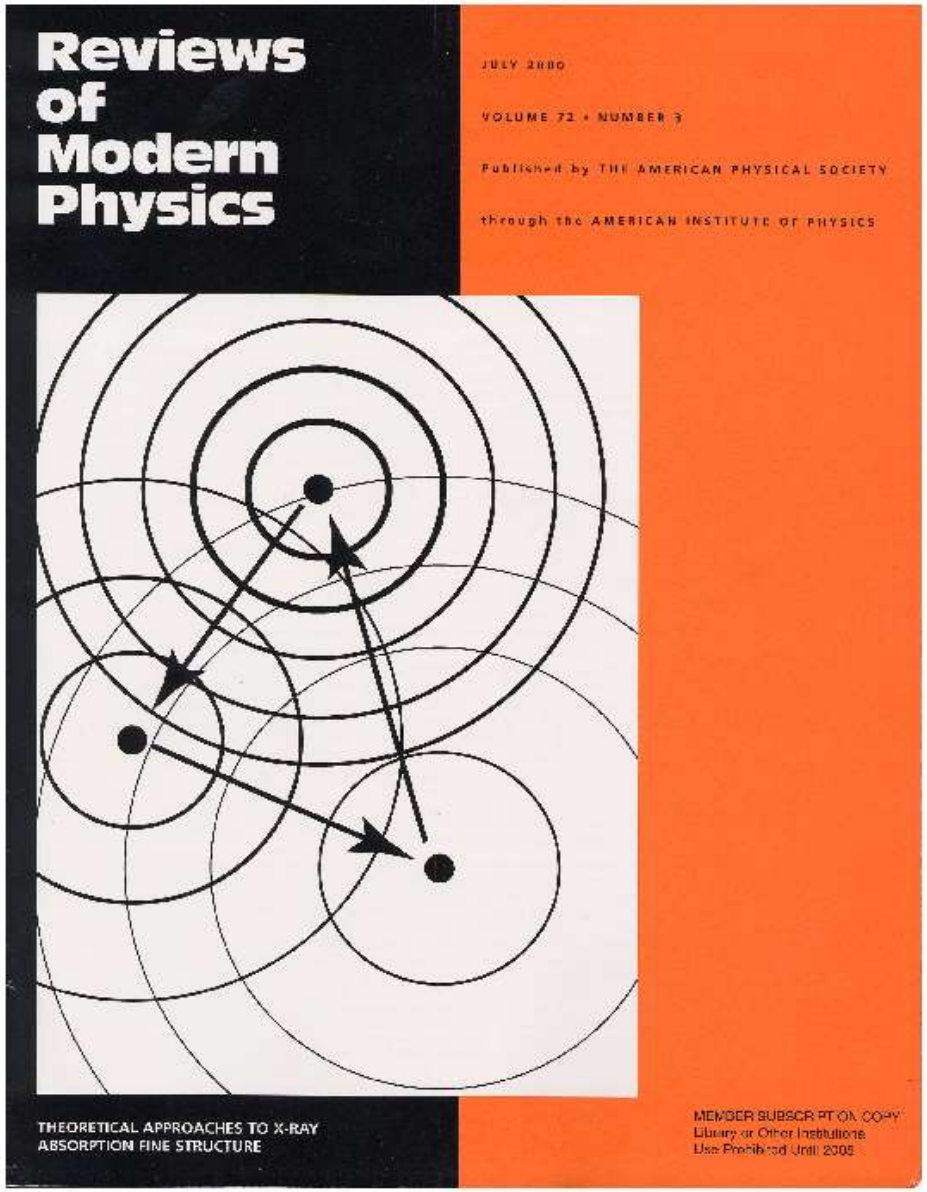
Example: Accurate distances R_{nn} from XAFS

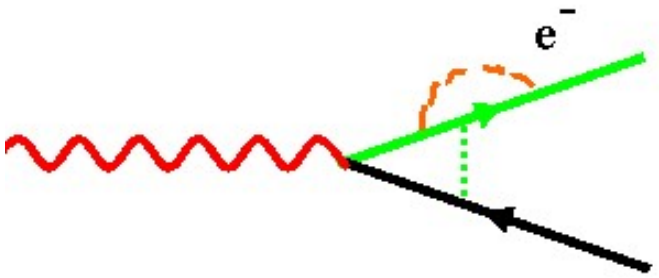
Phase Corrected XAFS Fourier Transform



Partial answer to Hans Bethe: "XAFS can be easier to calculate than to measure - ***IF*** **the structure is known"** JJR

Review: 30 yrs of developments in XAFS





FEFF

J. J. Rehr & R.C. Albers

Rev. Mod. Phys. **72**, 621 (2000)

http://feff.phys.washington.edu

Validation with Expt: IFEFFIT & ARTEMIS

J. Synchrotron Rad. **8**, 322 (2001)

IFEFFIT: interactive XAFS analysis and FEFF fitting

Matthew Newville*

Consortium for Advanced Radiation Sources, The University of Chicago, 5640 S Ellis Ave, Chicago, IL 60637, USA.
E-mail: newville@cars.uchicago.edu

J. Synchrotron Rad. **12**, 537 (2005)

ATHENA, ARTEMIS, HEPHAESTUS*: data analysis for X-ray absorption spectroscopy using *IFEFFIT

B. Ravel[a]* and M. Newville[b]

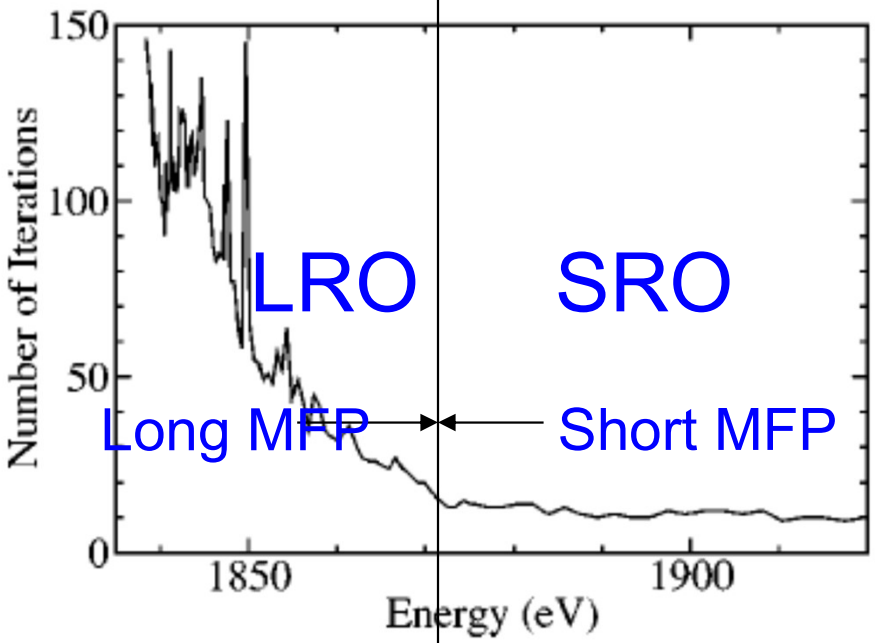
EXAFS vs XANES

PHYSICAL REVIEW B, VOLUME 65, 104107 (2002)

Parallel calculation of electron multiple scattering using Lanczos algorithms

A. L. Ankudinov,[1] C. E. Bouldin,[2] J. J. Rehr,[1] J. Sims,[2] and H. Hung[2]
[1]*Department of Physics, University of Washington, Seattle, Washington 98195*
[2]*National Institute of Standards and Technology, Gaithersburg, Maryland 20899*

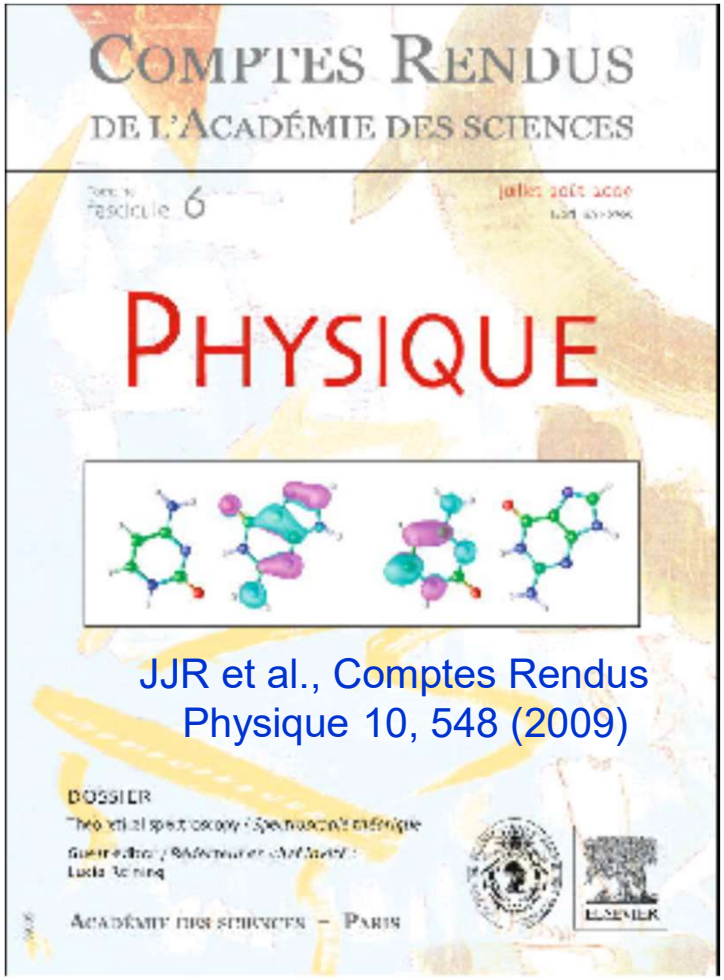
XANES EXAFS $E\text{-}E_0 > \omega_p \sim 30$ eV



$\lambda \sim k/[\Gamma + |\ \mathrm{Im}\ \Sigma\ |]$
$\sim [\ 5 - 20\ ]$ Å

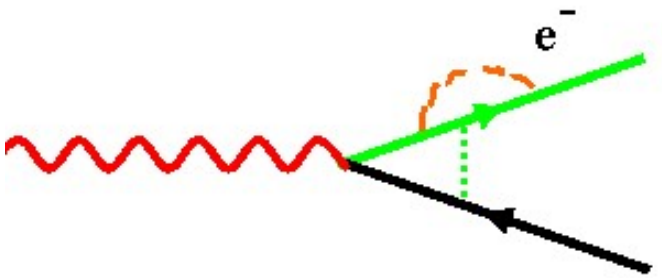
FMS Path expansion

Improved theory: Parameter free XAFS & XANES



JJR et al., Comptes Rendus Physique 10, 548 (2009)

Ab initio theory with *NO* adjustable parameters



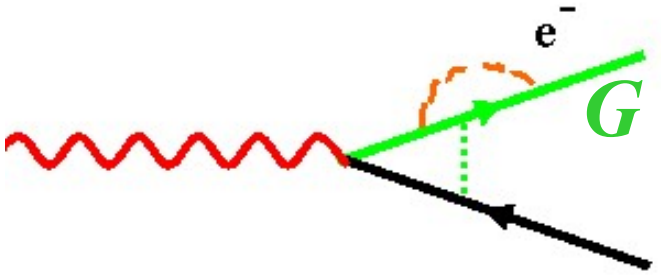
Phys. Chem. Chem. Phys. 12, 5503–5513 (2010)

PERSPECTIVE www.rsc.org/pccp | Physical Chemistry Chemical Physics

Parameter-free calculations of X-ray spectra with FEFF9

John J. Rehr,*[a] Joshua J. Kas,[a] Fernando D. Vila,[a] Micah P. Prange[bc] and Kevin Jorissen[a]

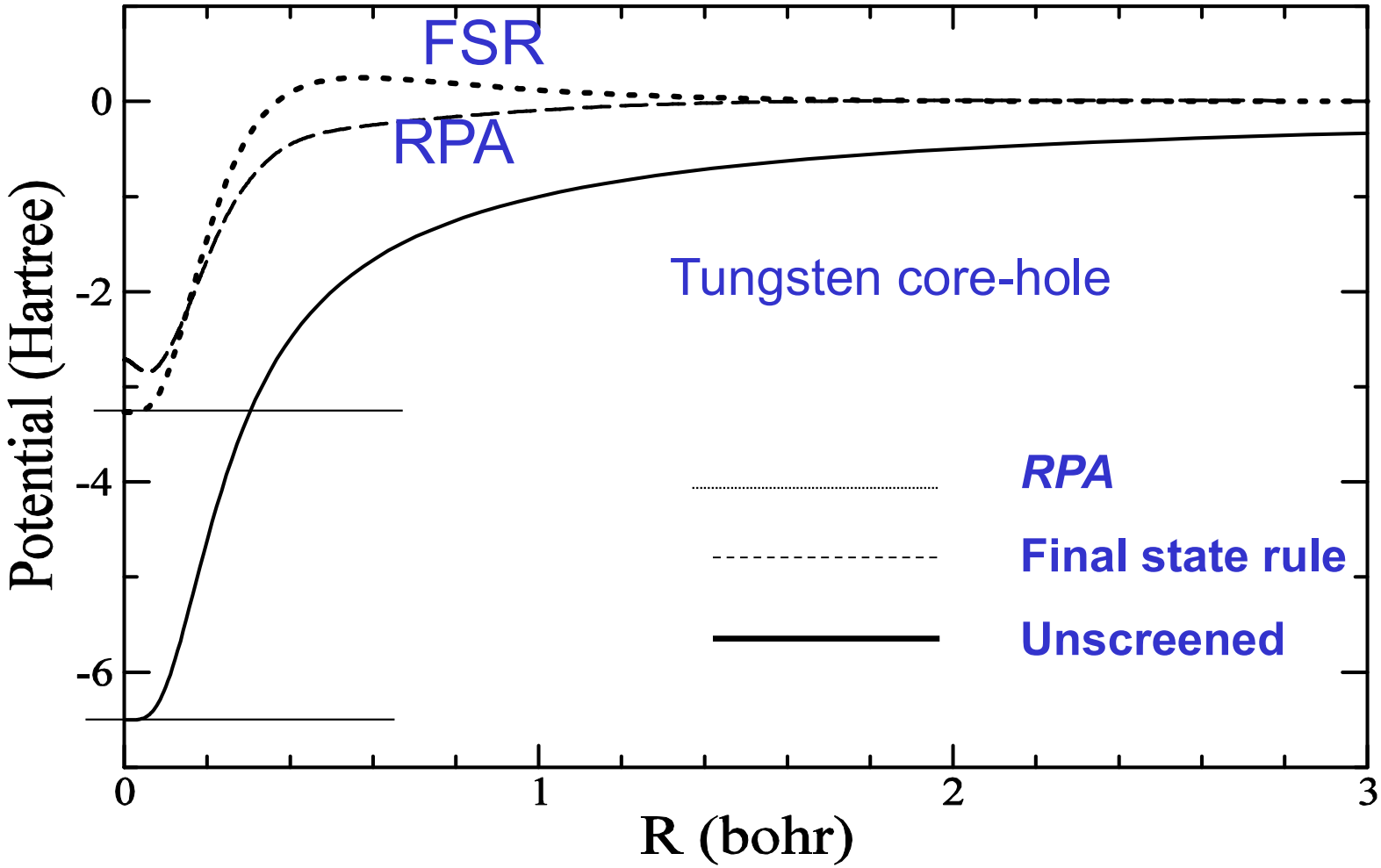
Ingredients in *ab initio* XAS theory



$$\mu = \mu_0(1 + \chi)$$

✓ Green's fn $G_{LR,L'R'}$	Separable propagators
✓ Atomic data $Z < 138$	Relativistic Dirac-Fock
✓ Scattering potentials $V(r)$	SCF, all-electron
✓ RPA Core-hole V_c	Screening, excitonic effects
✓ HL Self-energy $\Sigma(E)$	Mean-free path, energy shifts
✓ Ab initio Debye-Waller σ^2	Thermal vibrations
✓ Many body factor $S_0^2(E)$	Multi-electron excitations

RPA Screened core-hole potential ✓



RPA improves on final state rule, half-core hole, etc

.

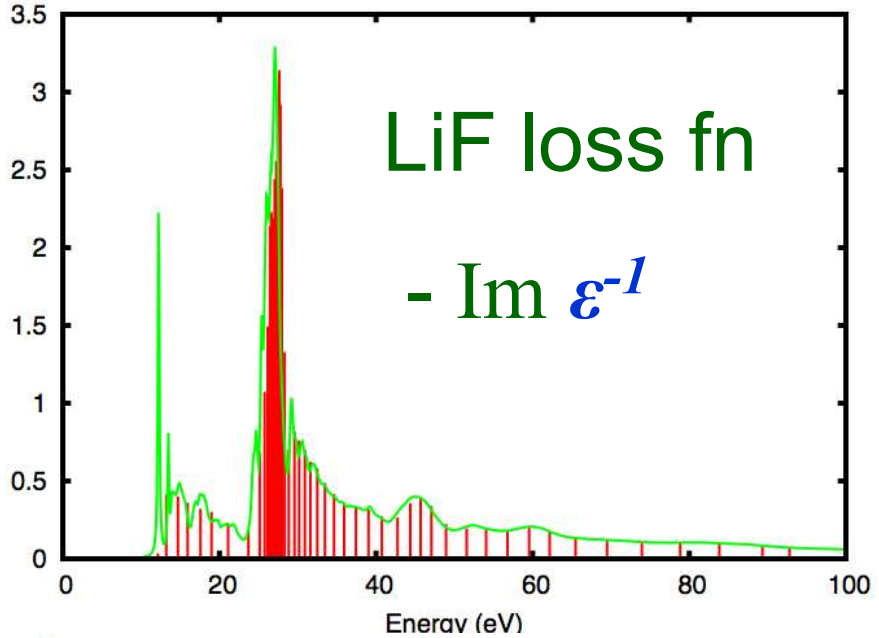
Many-pole GW Self-energy $\Sigma(E)$* ✓

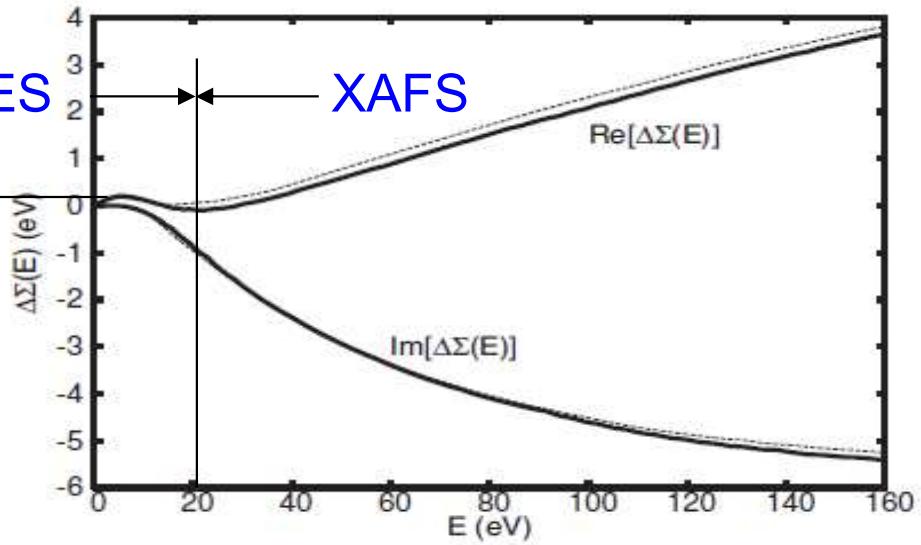
Efficient ~ GW approximation for self-energy Σ & mean free path λ

Sum of HL plasmon-pole models matched to loss function

$$\Sigma(E) = iGW = \Sigma' - i\,\Gamma$$

$$\varepsilon_2(\omega) = \frac{4\pi}{V}\mathrm{Im}\int \mathrm{d}r\mathrm{d}r'\,\mathrm{Tr}[d\chi(r,r';\omega)d^{\dagger}].$$





*J.J. Kas et. al, Phys Rev B **76**, 195116 (2007)

Ab initio Debye Waller factors ✓

An Initio Determination of Extended X-Ray Absorption Fine Structure Debye-Waller Factors*

$e^{-2\sigma^2 k^2}$

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H. H. Rossner and H. J. Krappe
Hahn-Meitner-Institut Berlin, Glienicker Strasse 100, D-14109 Berlin, Germany

(Dated: August 23, 2005)

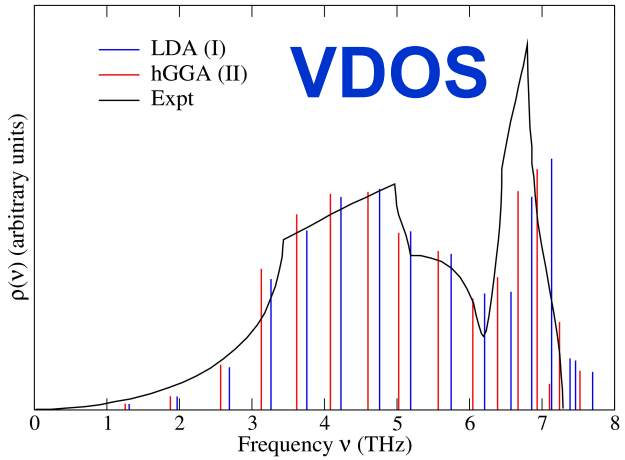
$$\sigma^2 = \frac{\hbar}{\mu_i}\int_0^\infty \rho(\omega^2)\coth\frac{\beta\hbar\omega}{2}\,d\omega$$

$$\rho(\omega^2) = \langle Q_i \,|\, \delta(\omega^2 - D) \,|\, Q_i \rangle$$

$$= \{6 - \text{step Lanczos recursion}\}$$

Many pole model for phonons

D dynamical matrix < DFT codes
ABINIT or Quantum Espresso



*F. Vila et al., Phys. Rev. B **76,** 014301 (2007)

Anharmonic effects & cumulant expansion

PHYSICAL REVIEW B VOLUME 48, NUMBER 1 1 JULY 1993-I

Thermal expansion and x-ray-absorption fine-structure cumulants

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Effects of large vibrational disorder beyond MSRD

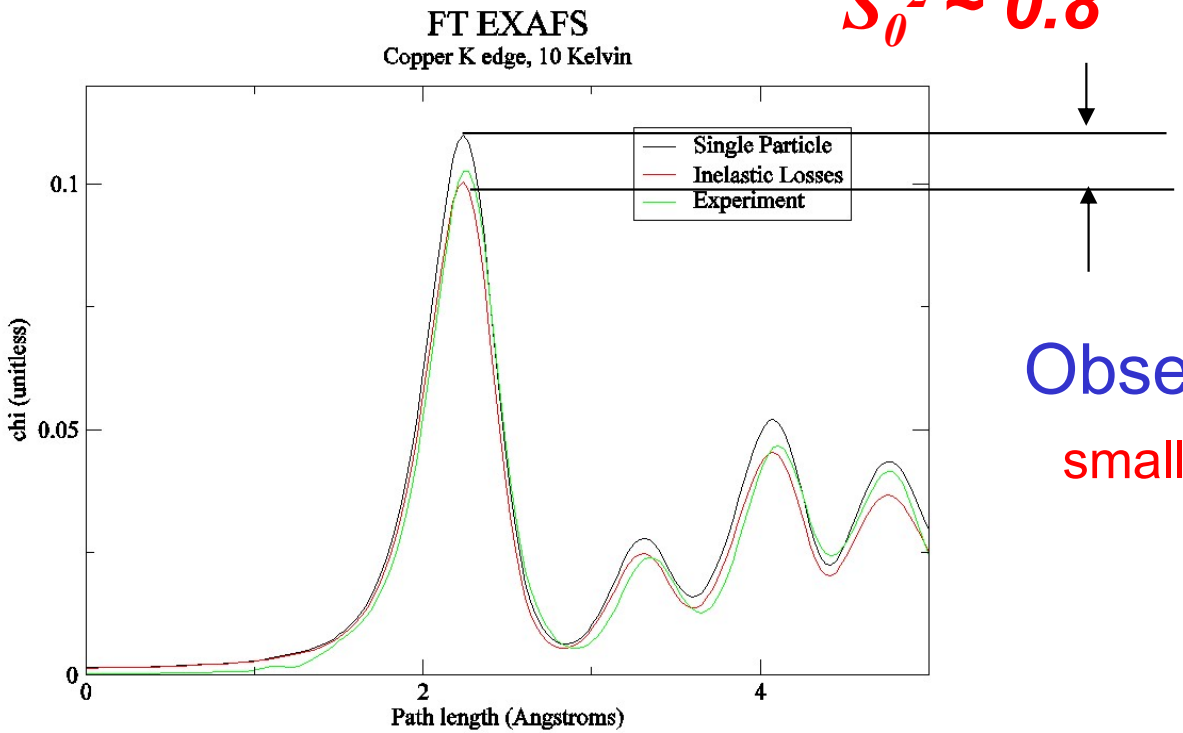
$$\langle e^{2ikR} \rangle = \exp[\sum_n \frac{C_n (2ik)^n}{n!}] \qquad C_n = \langle R^n \rangle_c$$

C_1 Thermal expansion, C_2 MSRD, C_3 skew …
& relations between cumulants C_1 C_2 & C_3

Last XAFS challenge: multi-electron excitations S_0^2

Q: How to treat S_0^2 theoretically?

$S_0^2 \sim 0.8$



Observed fine structure
smaller than QP theory

Shake-up & shake-off theory of S_0^2

PHYSICAL REVIEW B VOLUME 17, NUMBER 2 15 JANUARY 1978

Extended x-ray-absorption fine-structure amplitudes—Wave-function relaxation and chemical effects*

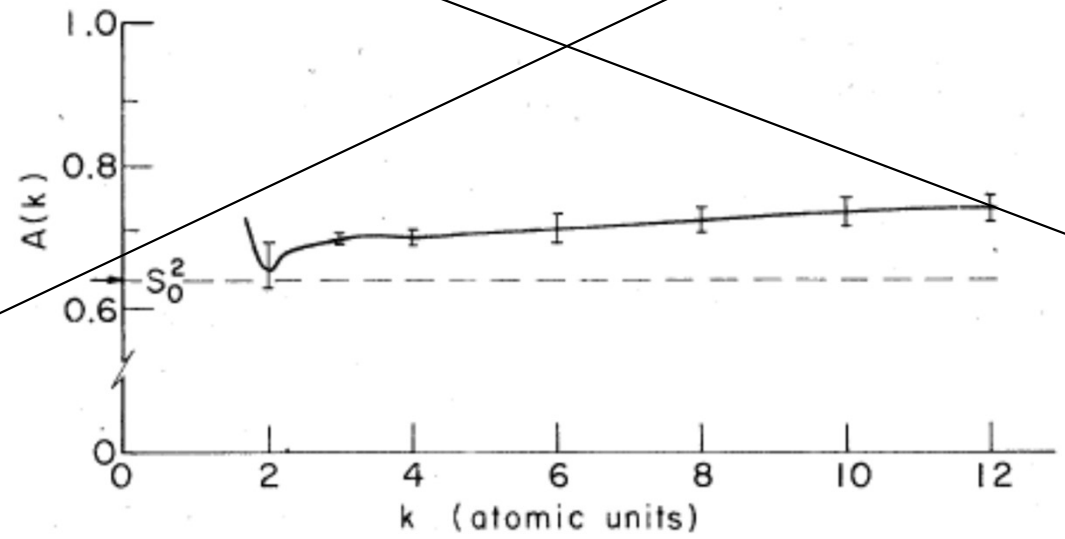
J. J. Rehr and E. A. Stern
Department of Physics, University of Washington, Seattle, Washington 98195

R. L. Martin† and E. R. Davidson
Department of Chemistry, University of Washington, Seattle, Washington 98195

Phasor sum of many-body overlap integrals

$$S_0^2 = \sum S_n^2 e^{2i(k_n - k)R}$$

Huge computational bottleneck



Suggestion: Approximate S_n^2 with XPS

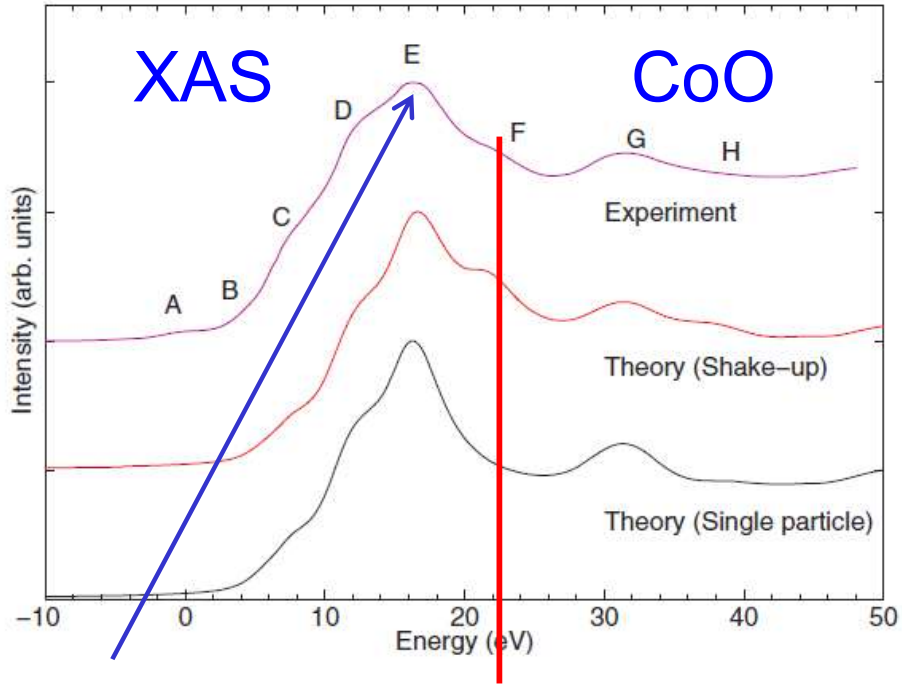
PHYSICAL REVIEW B 86, 165102 (2012)

K-edge x-ray absorption spectra in transition-metal oxides beyond the single-particle approximation: Shake-up many-body effects

M. Calandra,[1] J. P. Rueff,[2,3] C. Gougoussis,[1] D. Céolin,[2] M. Gorgoi,[4] S. Benedetti,[5] P. Torelli,[6] A. Shukla,[1] D. Chandesris,[7] and Ch. Brouder[1]

Convolution XPS * XAS

$$\sigma_{\mathrm{XAS}}(\omega) = \int d\epsilon\, \sigma_{\mathrm{XPS}}^{\mathrm{exp.}}(\epsilon)\sigma_{\mathrm{XAS}}^{\mathrm{sp}}(\omega - \epsilon$$

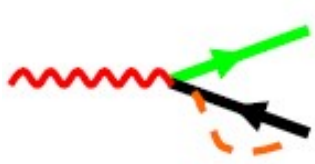


Reduction in peak height

Satellite peaks

Theory: XPS from core hole Green's function G_c

Intrinsic multi-electron excitations & plasmons due to suddenly turned-on core-hole:



Core-hole Green's function $G_c(\omega)$

Result: Convolution formula

Quasiparticle XAS

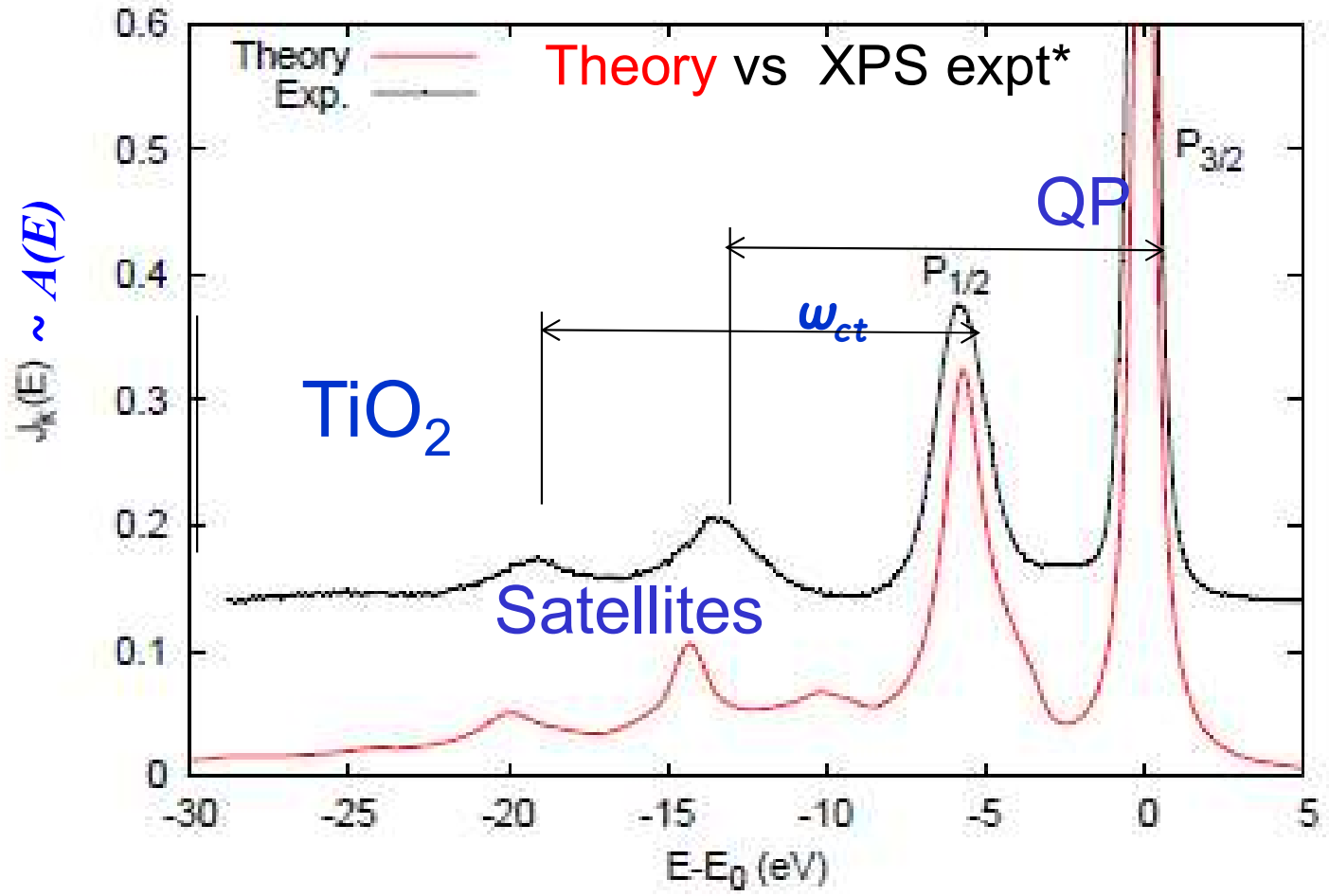
$$\mu(\omega) = \int_0^\infty d\omega' \, \tilde{A}(\omega, \omega') \mu_{qp}(\omega - \omega') \equiv \langle \mu_{qp}(\omega) \rangle$$

Core-hole spectral function – broadens & shifts XAS

$$A_c(\omega) = (1/\pi) \, |\mathrm{Im} \, G_c(\omega)|$$

Validation: Satellites in XPS

$$A_c(\omega) = (1/\pi)\,|\mathrm{Im}\,G_c(\omega)| \sim \text{XPS}\ J_k(E)$$



J. J. Kas, F. D. Vila, J. J. Rehr, and S. A. Chambers, PRB **91**, 121112(R) (2015)

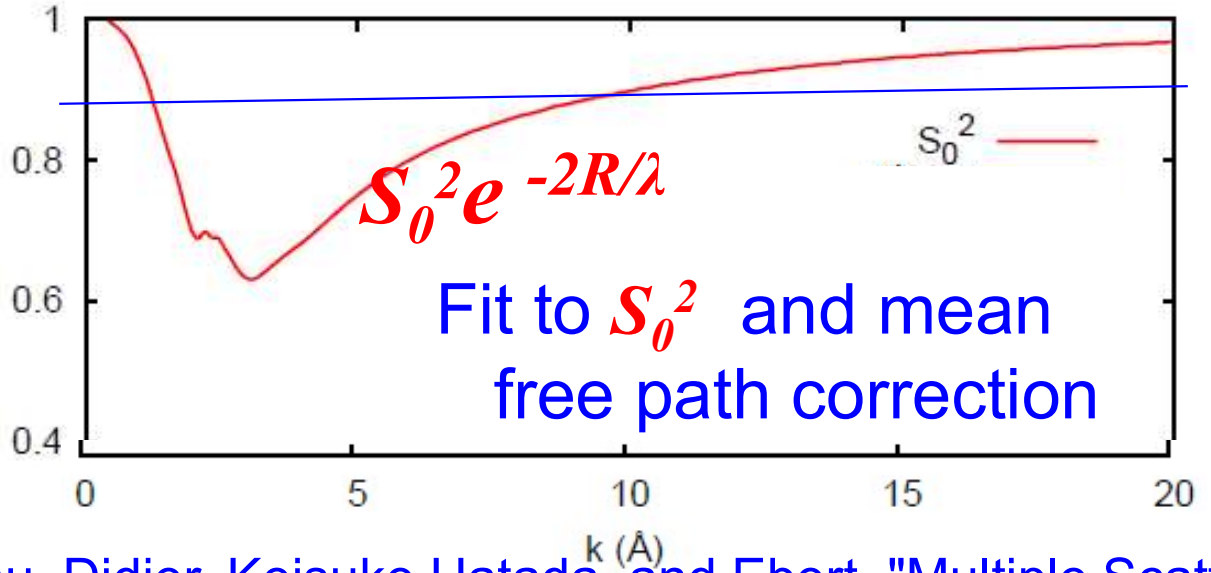
Application to S_0^2 in XAFS*

Cumulant approach for inelastic losses in x-ray spectra

John J. Rehr and Joshua J. Kas

Core-hole spectral function $A(\omega,\omega')$

$$S_0^2(R) = \int_0^{\omega} \mathrm{d}\omega' \tilde{A}(\omega,\omega') \mathrm{e}^{\mathrm{i}2[k(\omega-\omega')-k(\omega)]R}$$



*Ch. 27 in Sébilleau, Didier, Keisuke Hatada, and Ebert, "Multiple Scattering Theory for Spectroscopies." *Springer Proceedings in Physics* **204** (2018).

Improved theory: interference effects in S_0^2

Extrinsic + Intrinsic − 2 x Interference

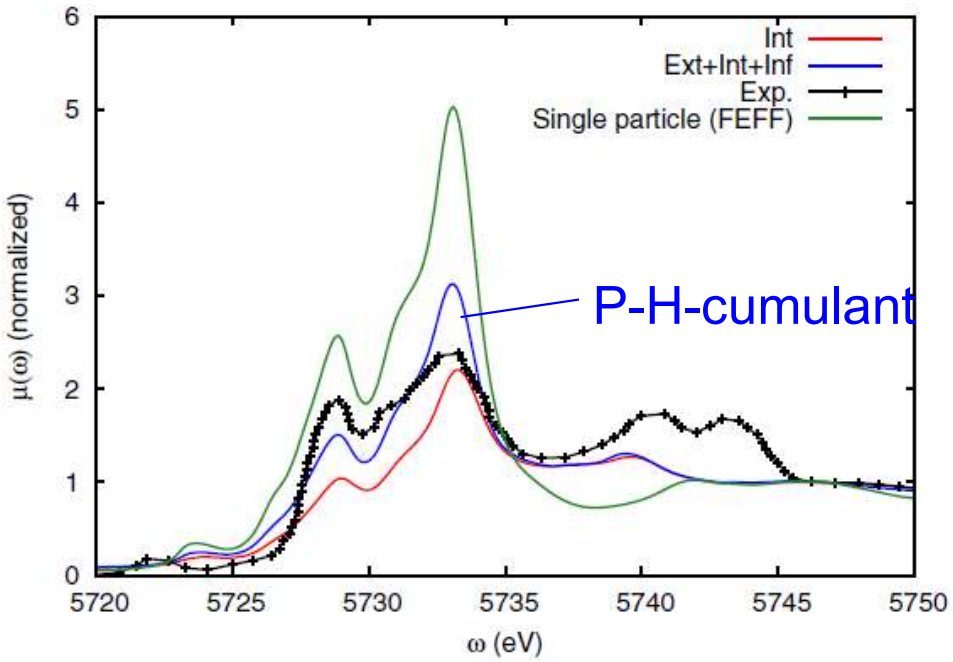
Interference reduces losses near E_0

All losses in convolution with *particle-hole spectral function*

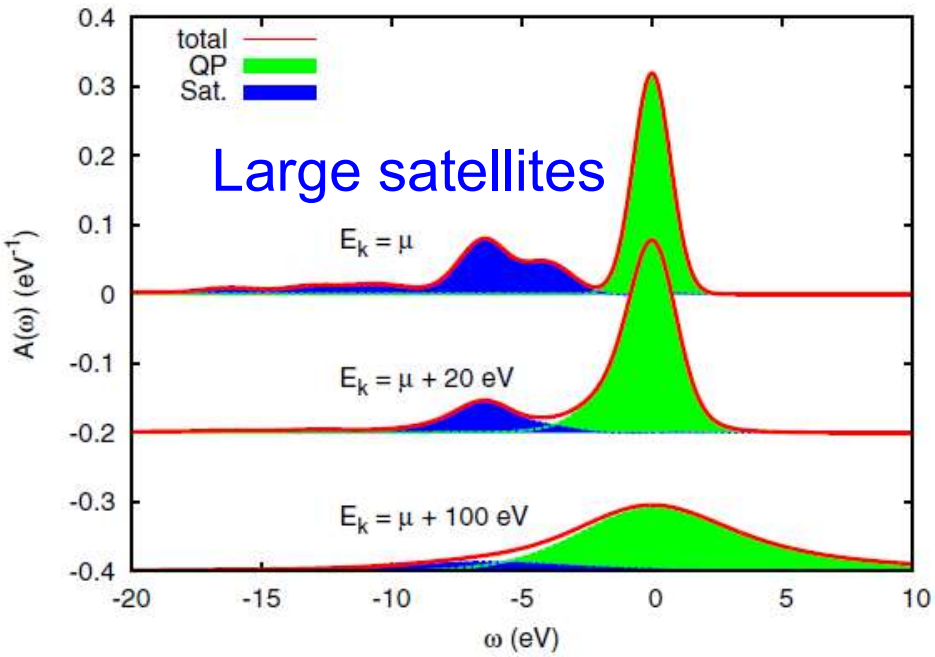
$$\mu(\omega) = \int d\omega' \, \tilde{A}_K(\omega') \mu_K(\omega - \omega')$$

Example: Particle-hole cumulant for CeO_2*

Ce L_3 XAS of CeO_2



Spectral function



Result: improves agreement in correlated systems

*J.J. Kas et al. Phys Rev B **94**, 035156 (2016)

III. Beyond XAFS - XANES & Related spectra

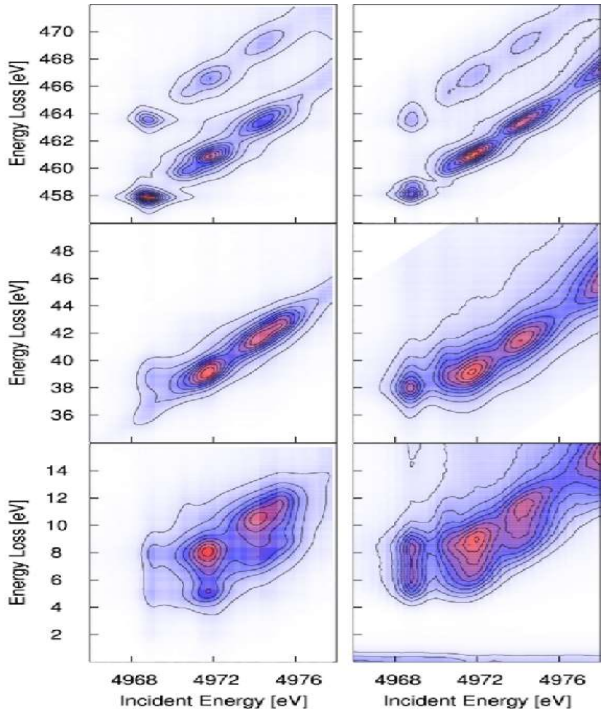
RIXS and NRIXS

PHYSICAL REVIEW B 83, 235114 (2011)

Real-space Green's function approach to resonant inelastic x-ray scattering

J. J. Kas,[1] J. J. Rehr,[1,*] J. A. Soininen,[2] and P. Glatzel[3]
[1]*Department of Physics, Box 351560, University of Washington, Seattle, Washington 98195-1560, USA*
[2]*Department of Physics, P.O. Box 64, University of Helsinki, FI-00014 Helsinki, Finland*
[3]*European Synchrotron Radiation Facility, B.P. 220, F-38043 Grenoble, France*
(Received 21 January 2011; revised manuscript received 7 April 2011; published 8 June 2011)

We present an *ab initio* theory of core and valence resonant inelastic x-ray scattering (RIXS) based on a real-space multiple-scattering Green's function formalism and a quasiboson model Hamiltonian. Simplifying assumptions are made that lead to an approximation of the RIXS spectrum in terms of a convolution of an effective x-ray absorption signal with the x-ray emission cross section. Additional many-body corrections are incorporated in terms of an effective energy-dependent spectral function. Example calculations of RIXS are found to give qualitative agreement with experimental data. Our approach also yields simulations of lifetime-broadening suppressed x-ray absorption, as observed in high-energy resolution fluorescence detection experiment. Finally, possible improvements to our approach are briefly discussed.



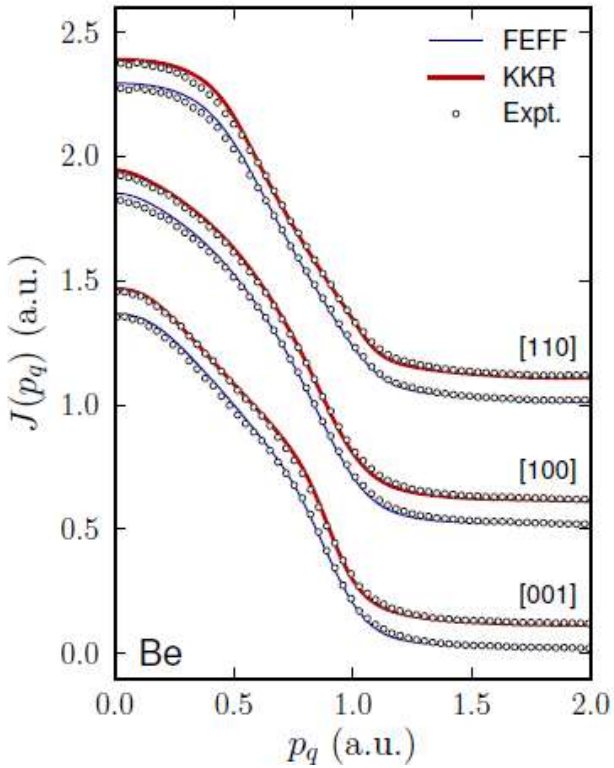
Compton, $S(\boldsymbol{q},\omega)$

PHYSICAL REVIEW B 85, 115135 (2012)

Real-space Green's function calculations of Compton profiles

Brian A. Mattern, Gerald T. Seidler,[+] Joshua J. Kas, Joseph I. Pacold, and John J. Rehr
Department of Physics, University of Washington, Seattle, Washington 98195-1560, USA
(Received 2 February 2012; revised manuscript received 16 March 2012; published 29 March 2012)

We report the development of a first-principles, real-space Green's function method for calculation of Compton profiles in the impulse approximation. For crystalline Be, we find excellent agreement with prior theoretical treatments requiring periodicity, with prior experimental measurements of the Compton profile, and with present measurements of the dynamical structure factor via nonresonant inelastic x-ray scattering (often also called x-ray Thomson scattering in the plasma physics community). We also find good agreement with prior experimental results for the Compton profile of Cu. This approach can be extended to disordered and very high-temperature systems, such as "warm dense matter," where theories presently used for the interpretation of inelastic x-ray scattering include condensed phase effects only at a perturbative level.



High temperature and extreme conditions

PRL 119, 176403 (2017) PHYSICAL REVIEW LETTERS

Finite Temperature Green's Function Approach for Excited State and Thermodynamic Properties of Cool to Warm Dense Matter

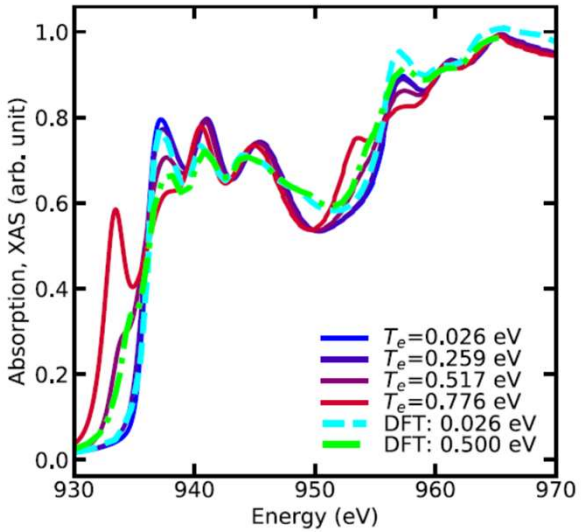
J. J. Kas and J. J. Rehr

PHYSICAL REVIEW B **104**, 035144 (2021)

Real-space Green's function approach for x-ray spectra at high temperature

Tun S. Tan, J. J. Kas, and J. J. Rehr*

Finite T cumulant Green's function theory of XAS



Multiplet + Cumulant Approach

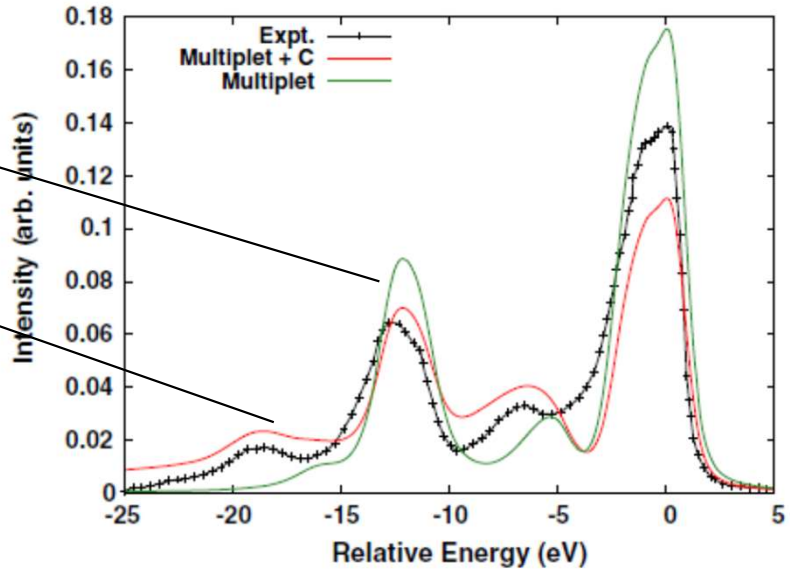
PHYSICAL REVIEW LETTERS **128,** 216401 (2022)

Ab Initio Multiplet-Plus-Cumulant Approach for Correlation Effects in X-Ray Photoelectron Spectroscopy

J. J. Kas,[1,*] J. J. Rehr,[1,2] and T. P. Devereaux[3,4]

Atomic multiplet theory ignores CT satellites

Multiplet (local) + Cumulant (long-range) approach yields multiplets, satellites and background spectrum



Full spectrum optical constants: UV-X-ray

Contents lists available at ScienceDirect

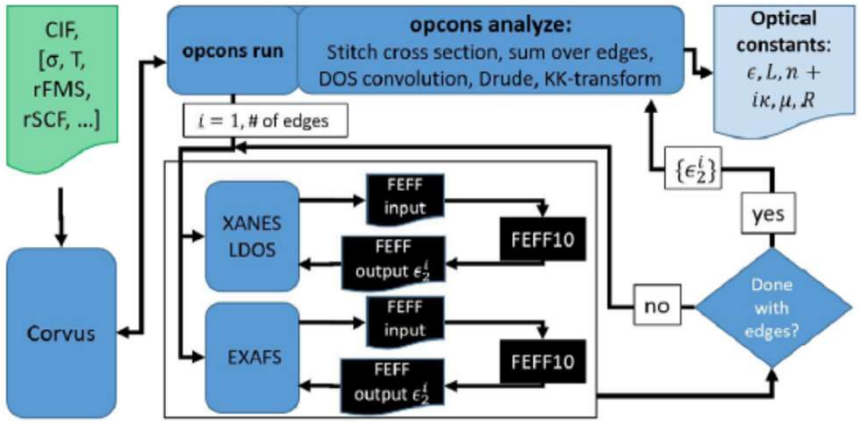
Computational Materials Science

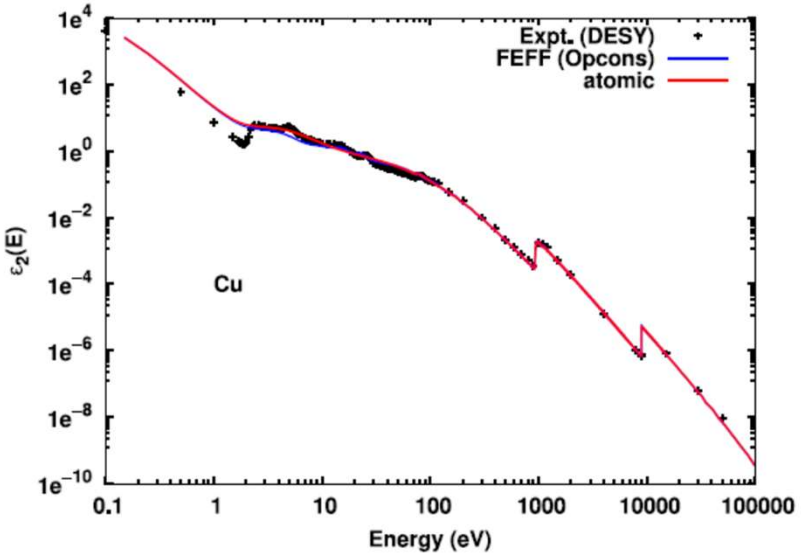
journal homepage: www.elsevier.com/locate/commatsci

Full spectrum optical constant interface to the Materials Project

J.J. Kas [a,b], F.D. Vila [a,b], C.D. Pemmaraju [b], M.P. Prange [c], K.A. Persson [d], R.X. Yang [d], J.J. Rehr [a,b,*]

Corvus Workflow Manager





High-throughput calculations for ML and AI

All ~10^5 materials & structures in MP Data base

Including feff.inp

SCIENTIFIC DATA

SCIENTIFIC DATA | 5:180151 | DOI: 10.1038/sdata.2018.151

Data Descriptor: High-throughput computational X-ray absorption spectroscopy

Kiran Mathew[1,*], Chen Zheng[2,*], Donald Winston[3], Chi Chen[2], Alan Dozier[4], John J. Rehr[5], Shyue Ping Ong[2] & Kristin A. Persson[1]

npj | Computational Materials

www.nature.com/npjcompumats

npj Computational Materials (2018) 4:12 ; doi:10.1038/s41524-018-0067-x Corrected: Author correction

ARTICLE OPEN

Automated generation and ensemble-learned matching of X-ray absorption spectra

Chen Zheng[1], Kiran Mathew[2], Chi Chen[1], Yiming Chen[1], Hanmei Tang[1], Alan Dozier[3], Joshua J. Kas[4], Fernando D. Vila[4], John J. Rehr[4], Louis F. J. Piper[5,6], Kristin A. Persson[2] and Shyue Ping Ong[1]

Improved XANES: Bethe-Salpeter Equation (BSE)

aka *Particle-Hole Green's function*

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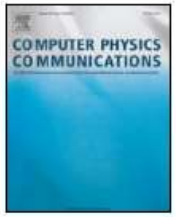


Ocean

Efficient implementation of core-excitation Bethe–Salpeter equation calculations

K. Gilmore [a,b,*], John Vinson [c], E.L. Shirley [c], D. Prendergast [d], C.D. Pemmaraju [d], J.J. Kas [e], F.D. Vila [e], J.J. Rehr [e]

IOP Publishing

Journal of Physics: Condensed Matter

J. Phys.: Condens. Matter 26 (2014) 363202 (24pp)

doi:10.1088/0953-8984/26/36/363202

Exciting

exciting: a full-potential all-electron package implementing density-functional theory and many-body perturbation theory

Andris Gulans[1], Stefan Kontur[1], Christian Meisenbichler[1], Dmitrii Nabok[1], Pasquale Pavone[1], Santiago Rigamonti[1], Stephan Sagmeister[2], Ute Werner[1] and Claudia Draxl[1,3]

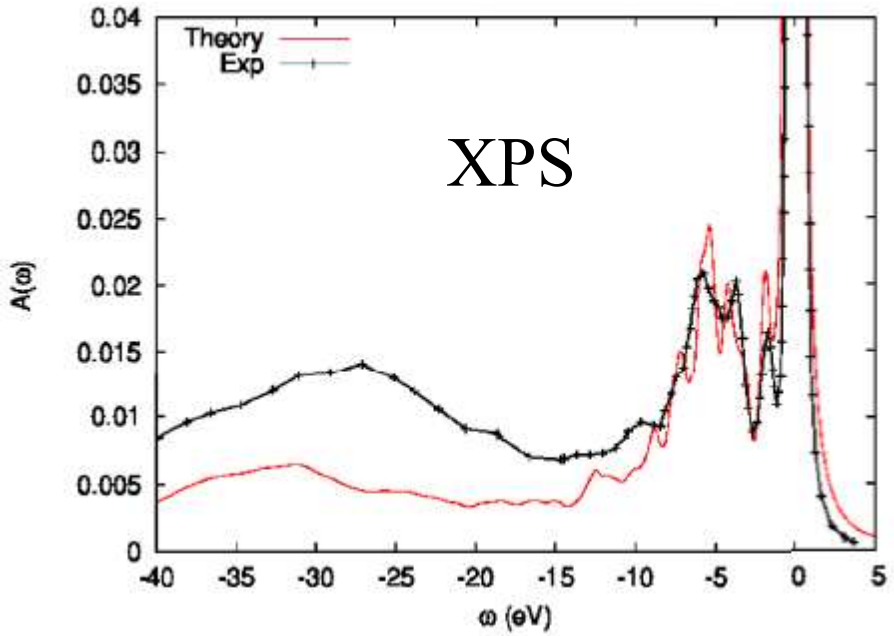
High accuracy XPS and XANES

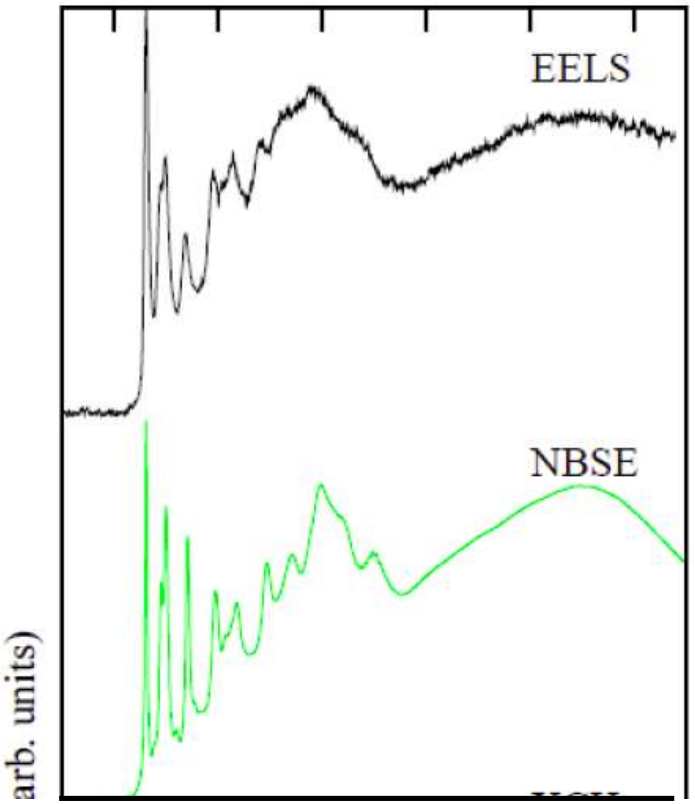
Phys. Rev. B **95**, 115112 (2017)

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

BSE ∗ Particle-hole spectral function





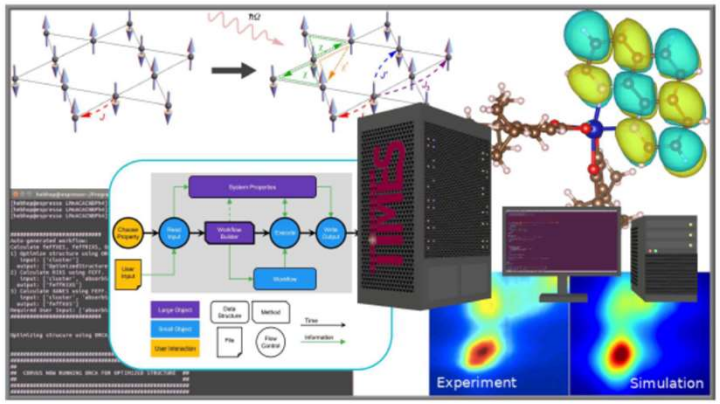
Next generation XAS software

TIMES@SLAC

FEFF10, Corvus, etc

U.S. Department of Energy: Office of Basic Energy Sciences
Materials Sciences & Engineering Division

Theory Institute for Materials and Energy Spectroscopies (TIMES)



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EOM-CC, Real-time, etc

July 2017

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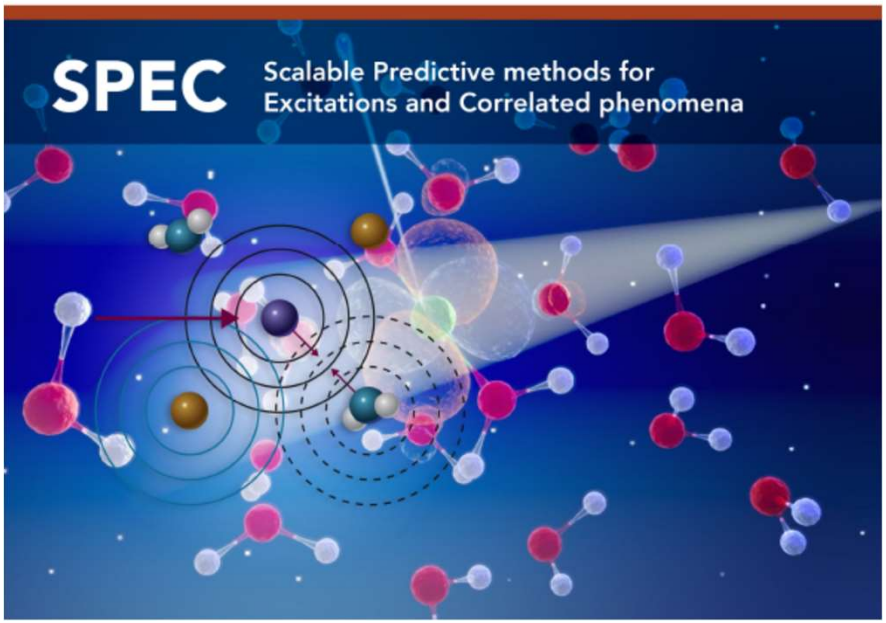

Conclusions

Are we there yet after 50 years ?

XAFS − Fairly high accuracy now possible with *ab initio* *GF* theory & automated codes FEFF10

XANES, XES, RIXS, OPCONS etc.

− High accuracy now possible with full potential BSE, DFT-MD and cumulant GF methods

NIRVANA: High accuracy Corvus workflows for full-spectrum XAS and related spectra may well be possible !

Acknowledgments – Sincere thanks to our students, postdocs & collaborators experimental colleagues, and especially the DOE for support

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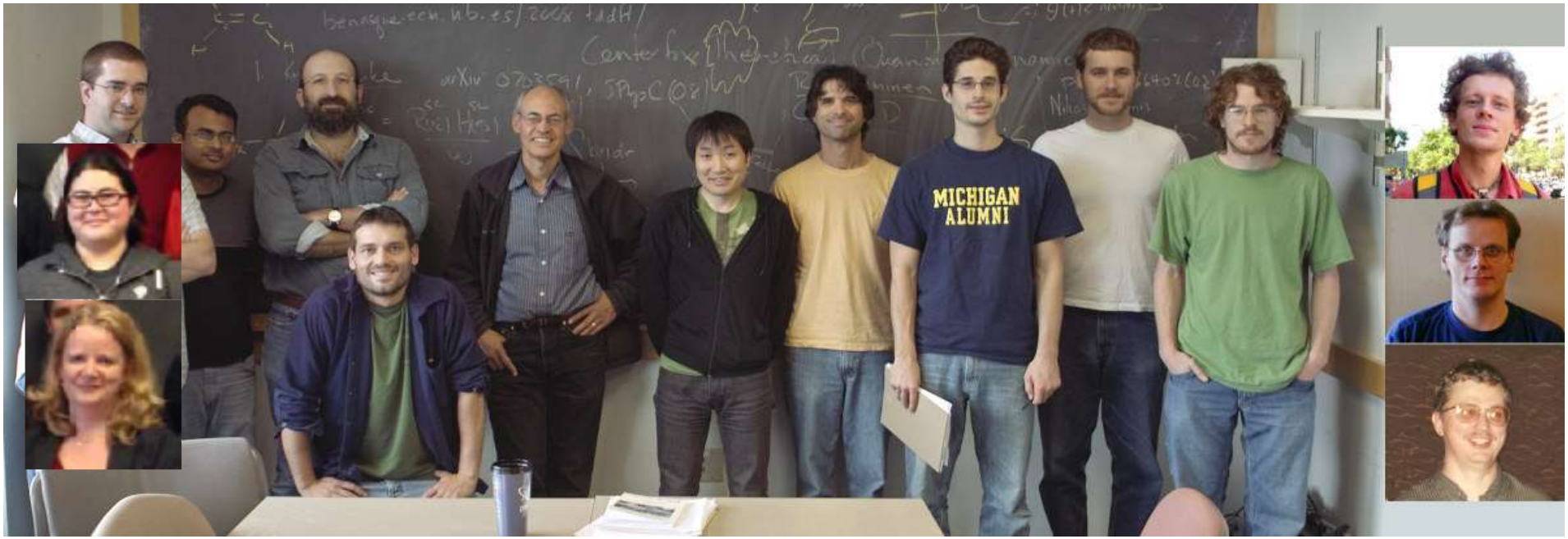

UNIVERSITY OF WASHINGTON TIMES Pacific Northwest NATIONAL LABORATORY SPEC U.S. DEPARTMENT OF ENERGY Office of Science