X-ray-Raman-scattering-based EXAFS beyond dipole limit

XAS and its Limitations

XRS

- **Inelastic X-ray scattering through XRS**
- **Advantage of XRS over XAS**
- **Discovery of XRS**
- **True potential and increasing use of XRS**

Application of XRS to EXAFS studies

- **Momentum-transfer dependence of XRS in EXAFS regime**
- **Non-Dipole contribution in case of XRS based EXAFS**
- **Complications in analysis of results**

Experimental Setup

- Source
- Monochromator
- Spectometer
- Detector
- Monitor
- Sample

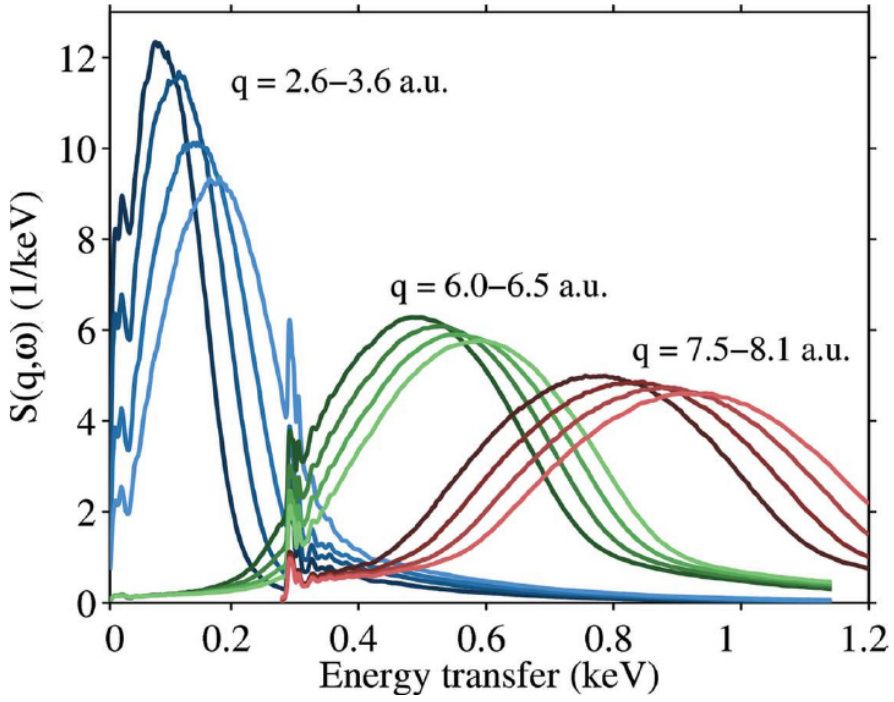
NRIXS

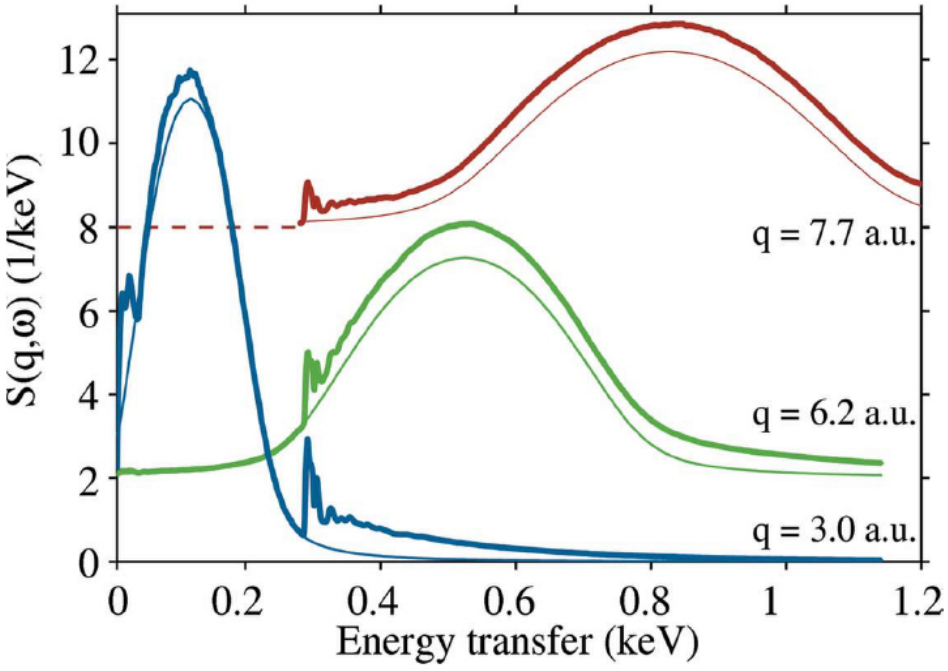
$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}\omega} = \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\mathrm{Th}} S(\mathbf{q},\omega),$$

Thomson Scattering $$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\mathrm{Th}} = r_{\mathrm{e}}^2 \frac{\omega_2}{\omega_1}(\hat{\mathbf{e}}_1 \cdot \hat{\mathbf{e}}_2)^2,$$

Structure Factor $$S(\mathbf{q},\omega) = \sum_F \left|\left\langle F\right| \sum_j \exp\left(i\mathbf{q}\cdot\mathbf{r}_j\right)\left|I\right\rangle\right|^2 \delta(\omega + E_I - E_F),$$

Dynamic Structure Factor





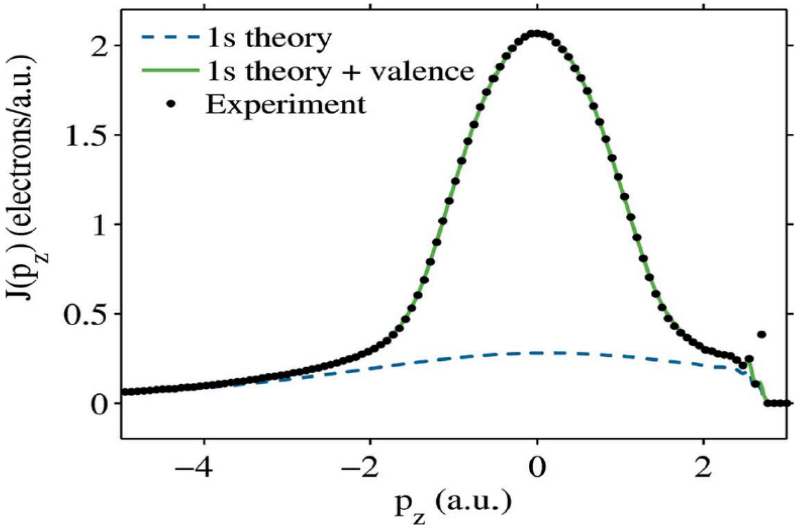
Data Analysis

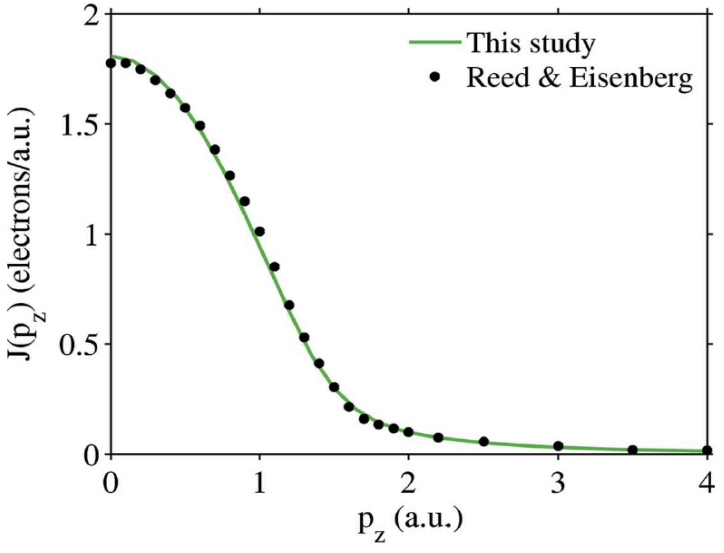
Compton Profile

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}\omega} = \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\mathrm{Th}} \frac{1}{q} J(p_z),$$

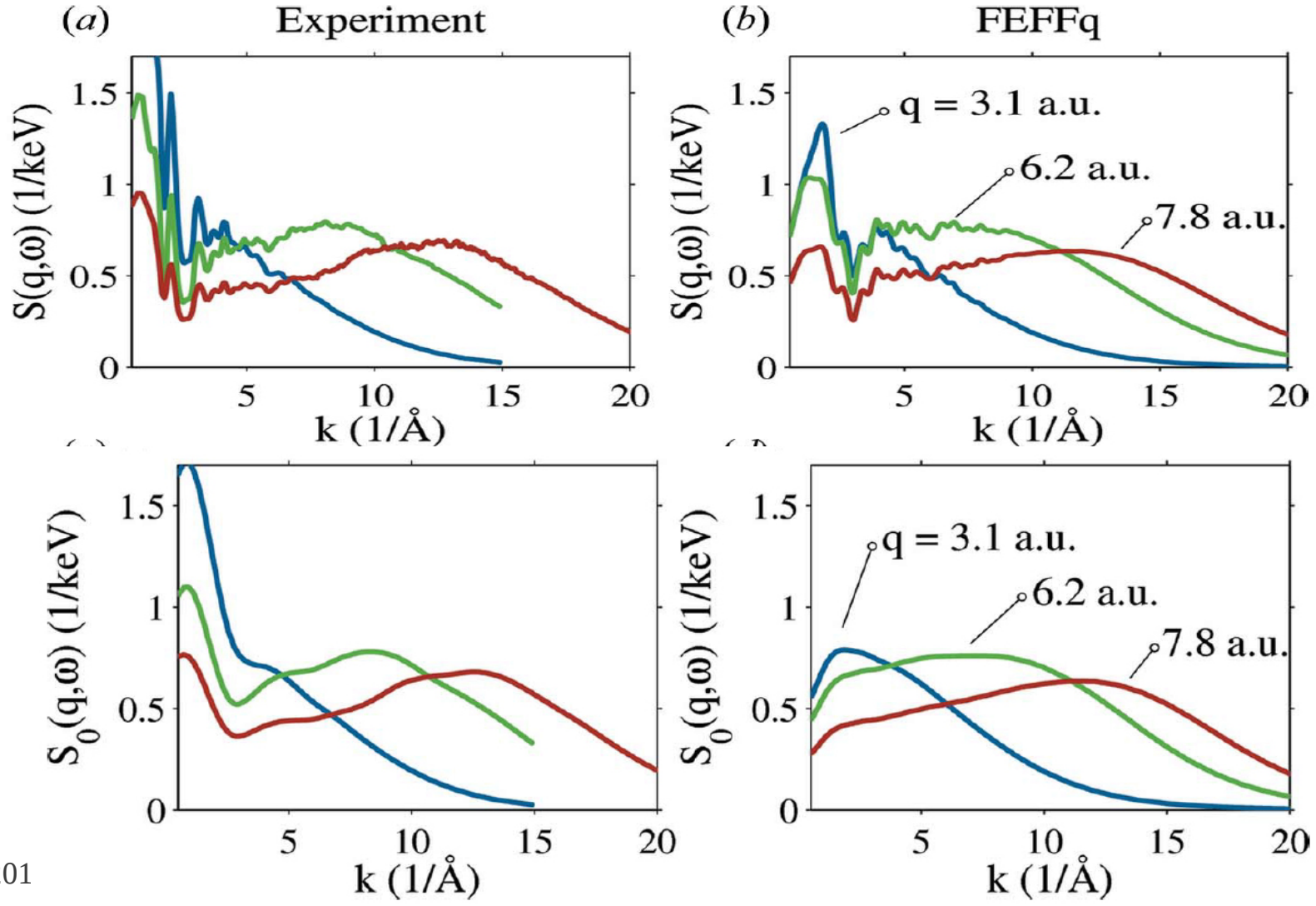
$$J(p_z) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} N(\mathbf{p})\,\mathrm{d}p_x\mathrm{d}p_y.$$

$$J(p_z) = (1/2)\int \mathrm{d}\Omega \int_{|p_z|}^{\infty} N(\mathbf{p})\,p\,\mathrm{d}p.$$





Core Electron Structure Factor



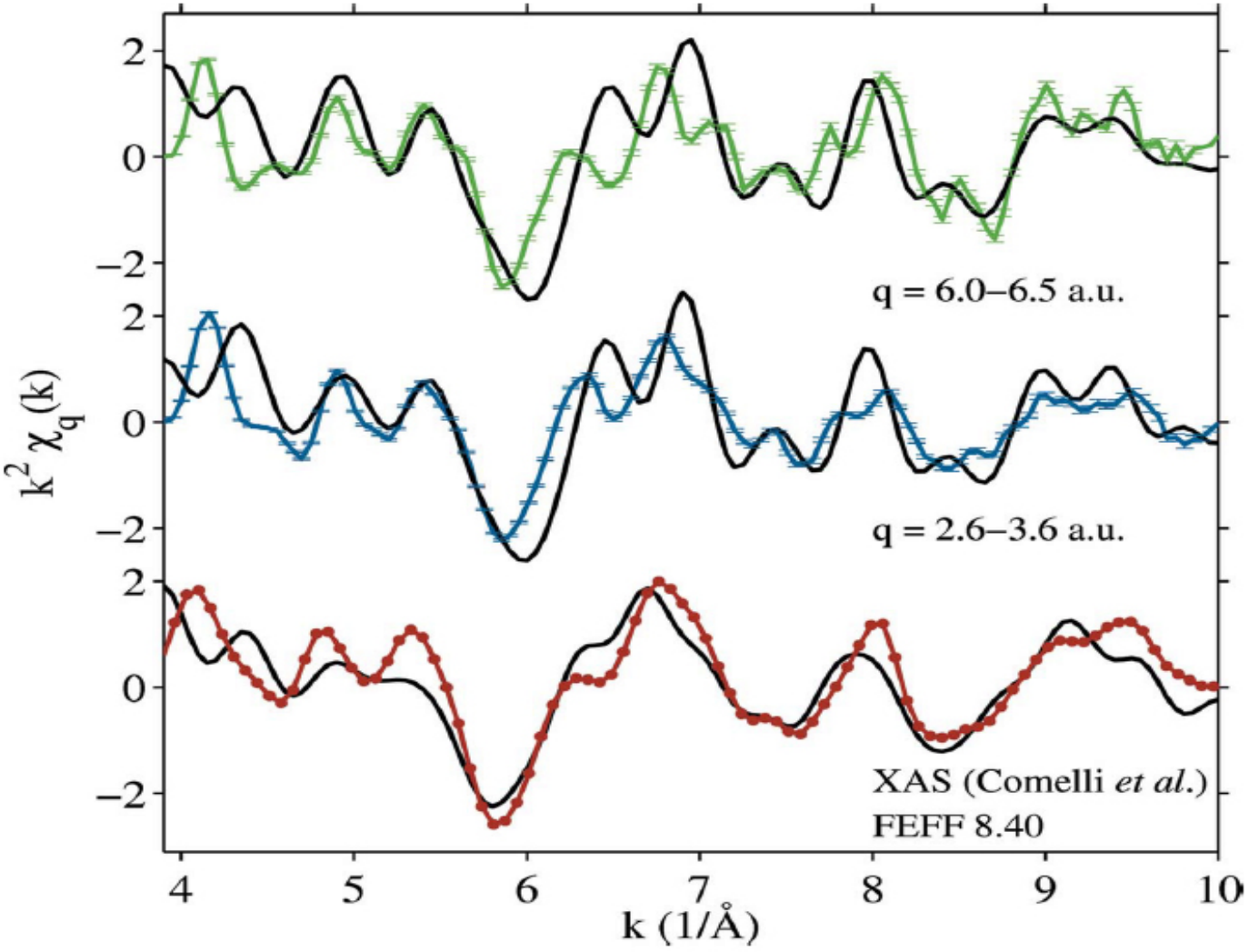
EXAFS Extraction

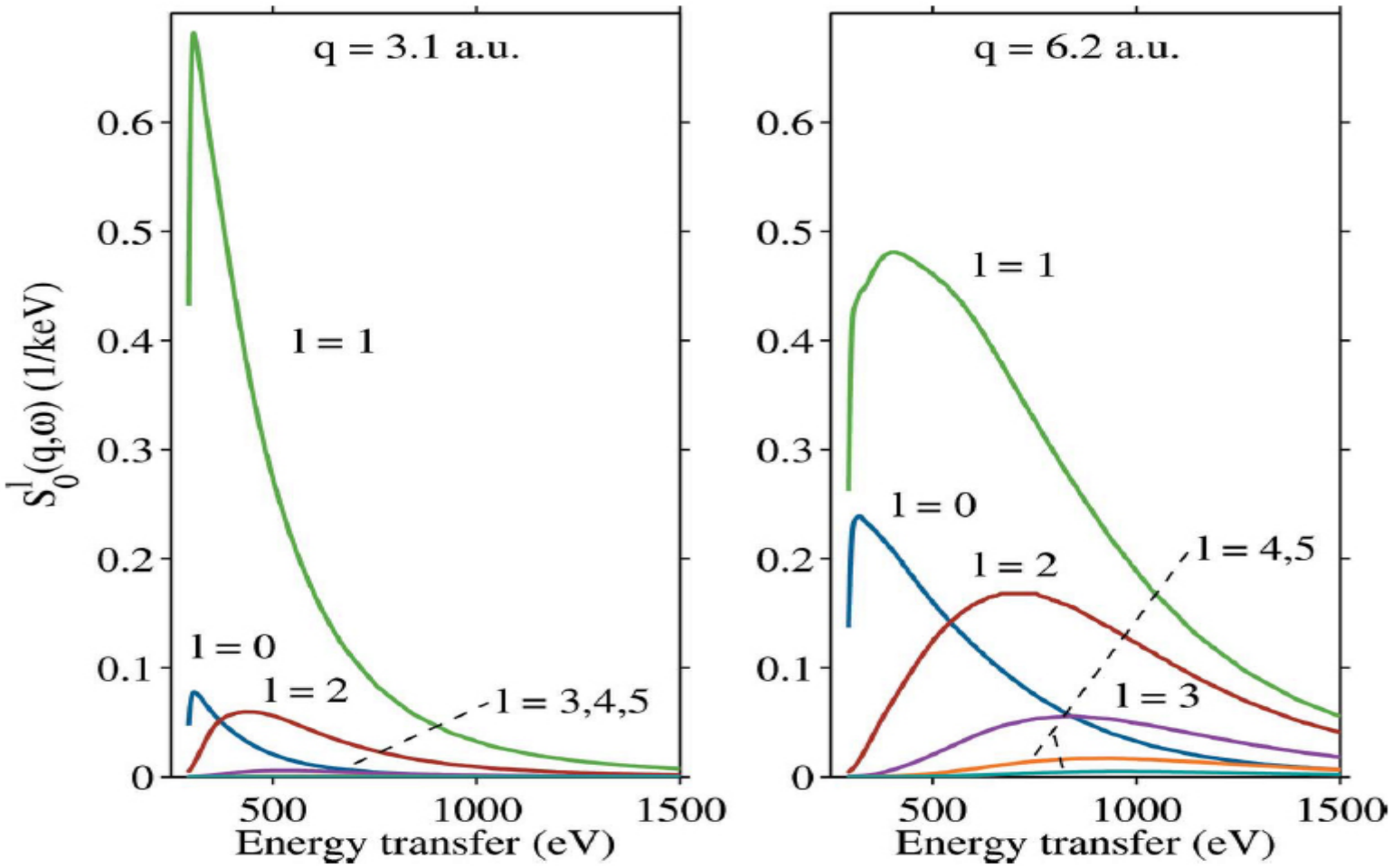
$$S(q,\omega) = \sum_l (2l+1)|M_l(q,\omega)|^2 \rho_l(\omega),$$

$$S(q,\omega) = S_0(q,\omega)\big[1+\chi_q(k)\big],$$

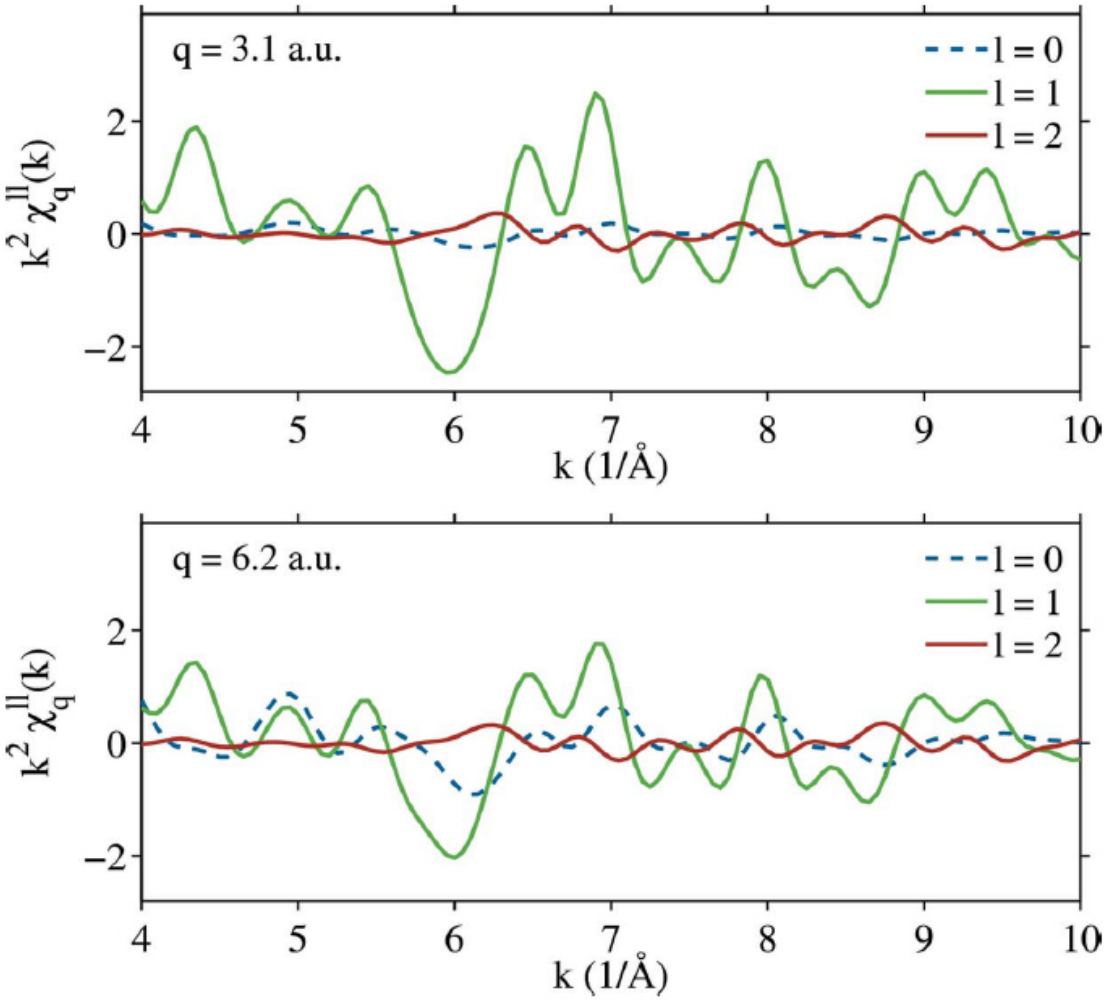
$$S_0(q,\omega) = \sum_l S_0^l(q,\omega), \qquad \chi_q(k) = \sum_{l,l'} \chi_q^{ll'}(k).$$

Results

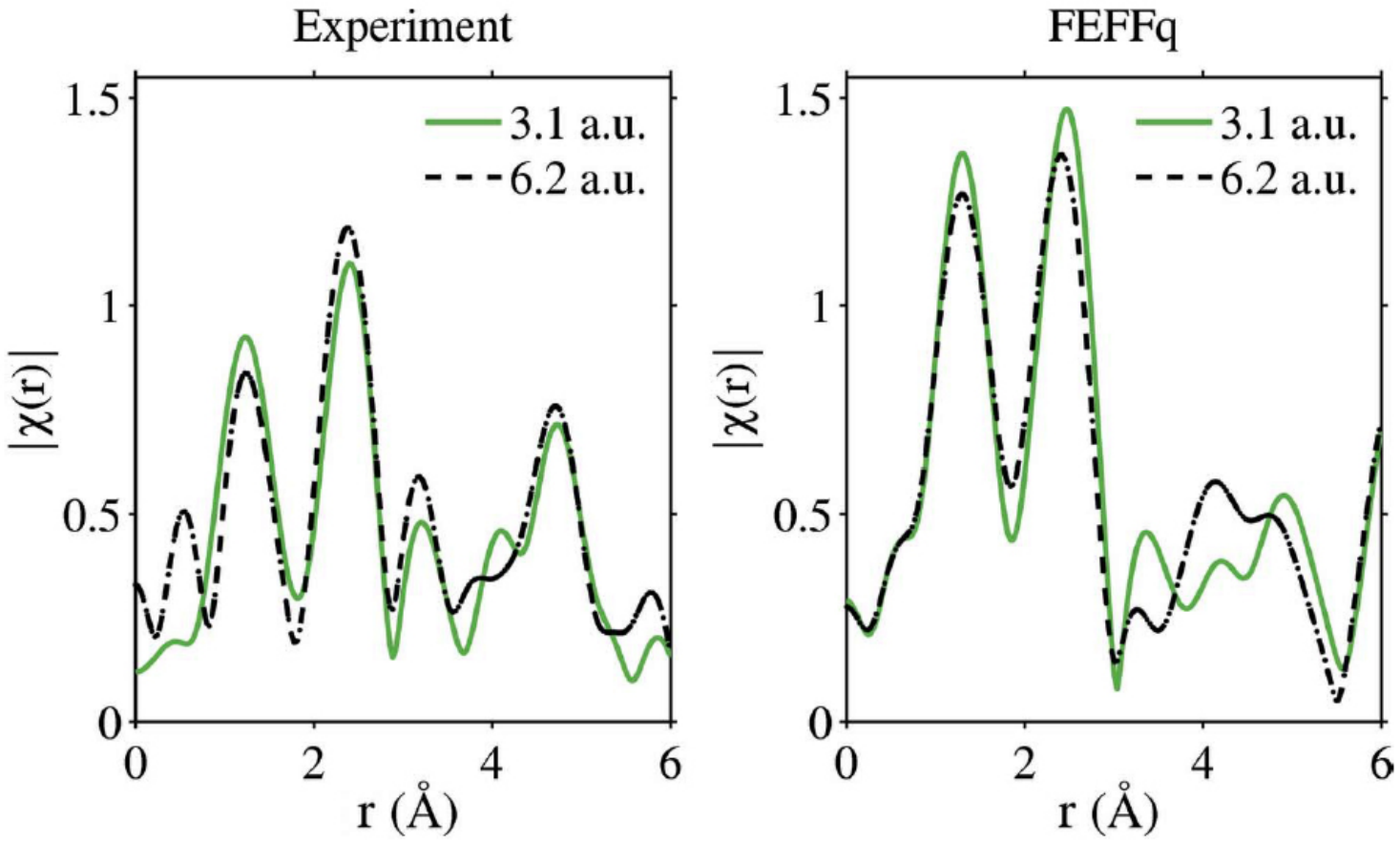

2
0
−2
q = 6.0–6.5 a.u.
q = 2.6–3.6 a.u.
XAS (Comelli *et al.*)
FEFF 8.40
$k^2 \chi_q(k)$
4
5
6
7
8
9
10
k (1/Å)



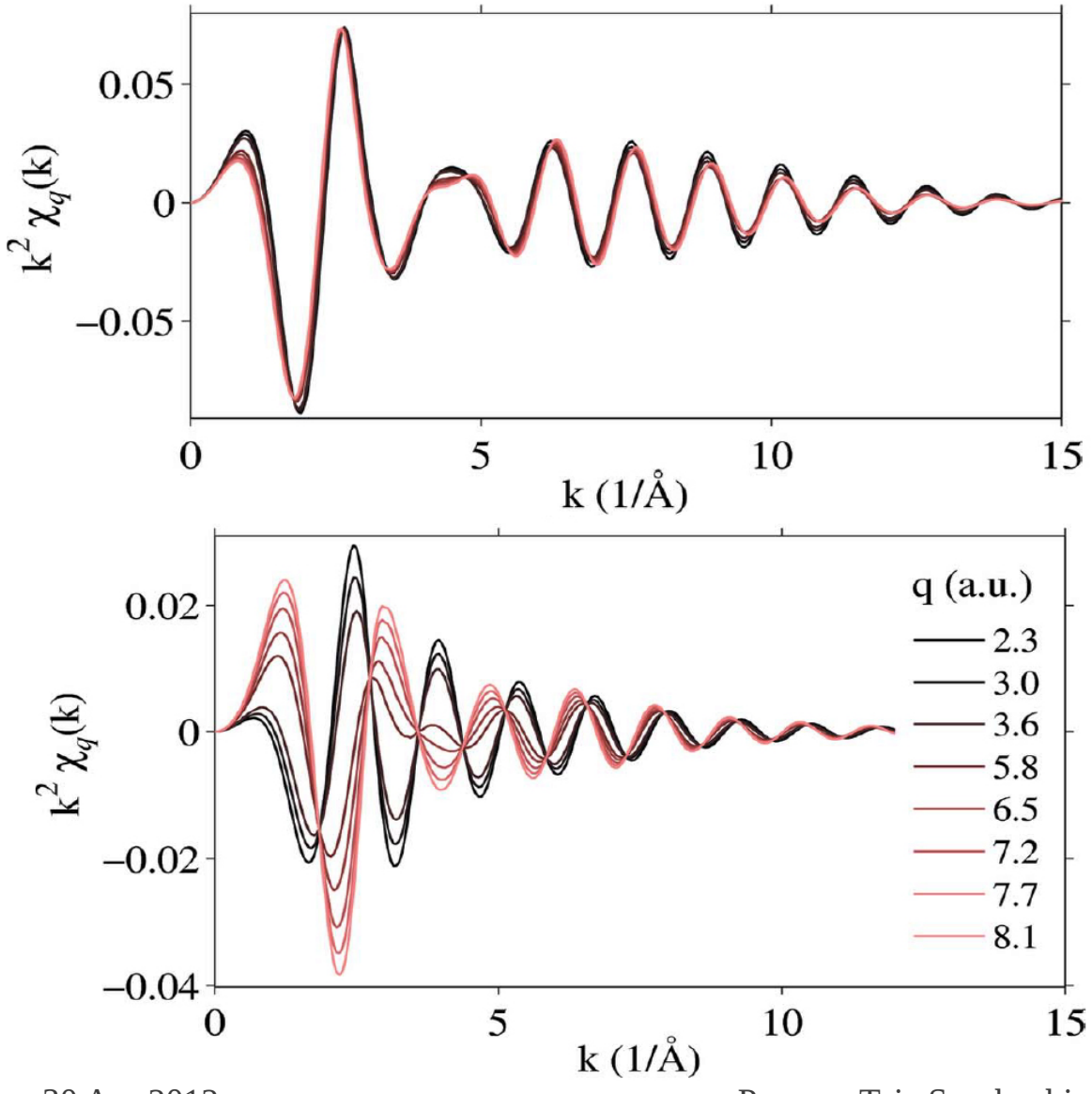
Angular Momentum contribution to carbon K-edge Structure factor



Angular momentum contributions to EXAFS calculations



Experiment and FEFFq calculations of Diamond for two momentum transfers



Momentum-transfer dependence of two triangular paths with equal half-path distance

Conclusions