Probing electron excitations with inelastic x-ray scattering spectroscopies

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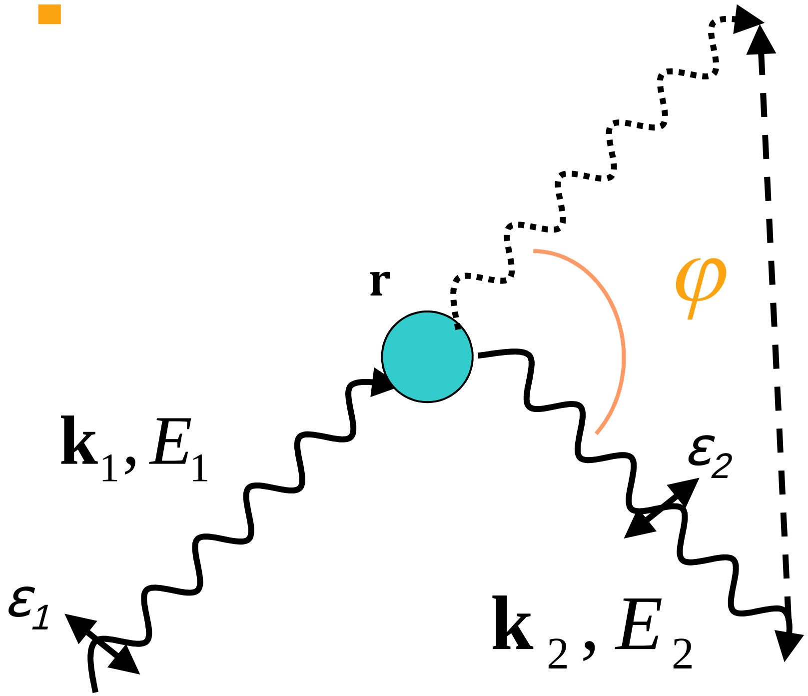
Benasque TDDFT workshop January 2012

Inelastic x-ray scattering



Dynamic structure factor S(Q,E)

$$S(\mathbf{Q},E) \propto \sum_f \left|\langle \Psi_f | e^{i\,\mathbf{Q}\cdot\mathbf{r}} | \Psi_i \rangle\right|^2 \delta\left(E_i - E_f + E\right)$$

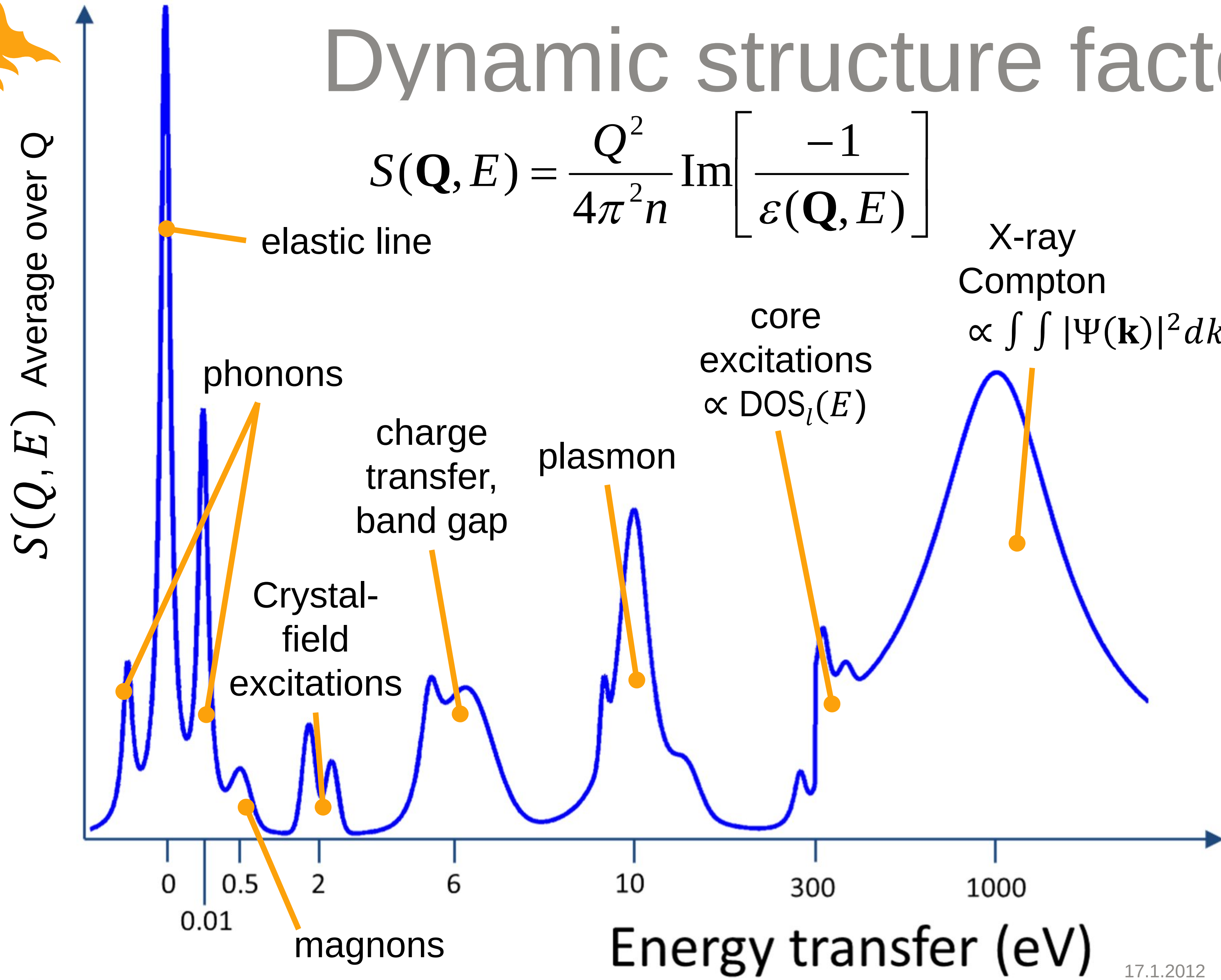
$$\boxed{\mathbf{Q} = \mathbf{k}_1 - \mathbf{k}_2}$$

$$S(\mathbf{Q},E) = \frac{Q^2}{4\pi^2 n}\,\mathrm{Im}\left[\frac{-1}{\varepsilon(\mathbf{Q},E)}\right]$$

Momentum transfer	$\mathbf{Q} = \mathbf{k}_1 - \mathbf{k}_2$
Energy transfer	$E = E_1 - E_2$

Dynamic structure factor

$$S(\mathbf{Q},E) = \frac{Q^2}{4\pi^2 n}\,\mathrm{Im}\left[\frac{-1}{\varepsilon(\mathbf{Q},E)}\right]$$



Outline

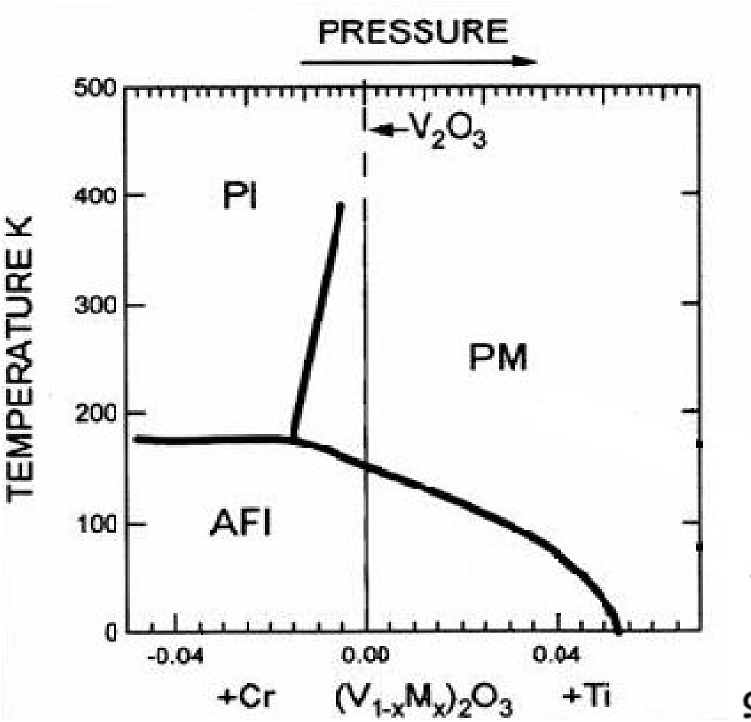
Part 1 Metal-to-insulator transition

Part 2 Plasmons and e-h continuum

Part 3 Double plasmons

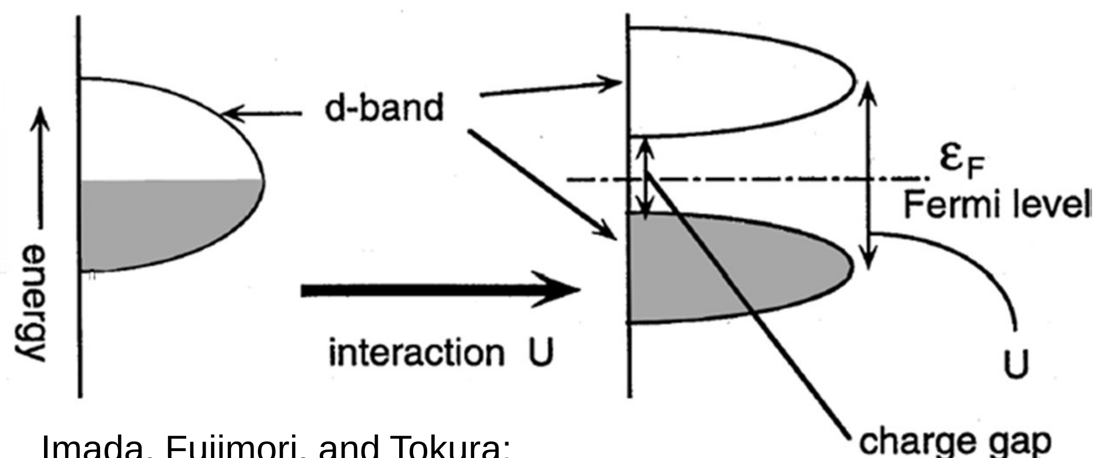
Part 4 Quasiparticle renormalisation

V_2O_3 and the Mott transition



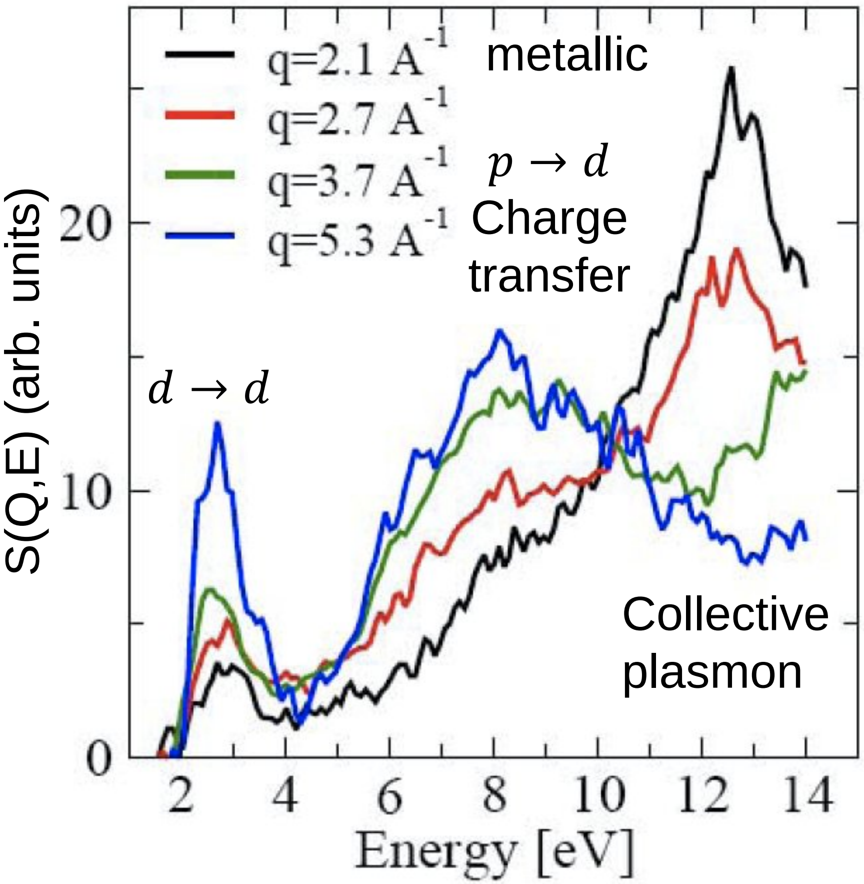
*McWhan et al., PRL **27** (1971)*

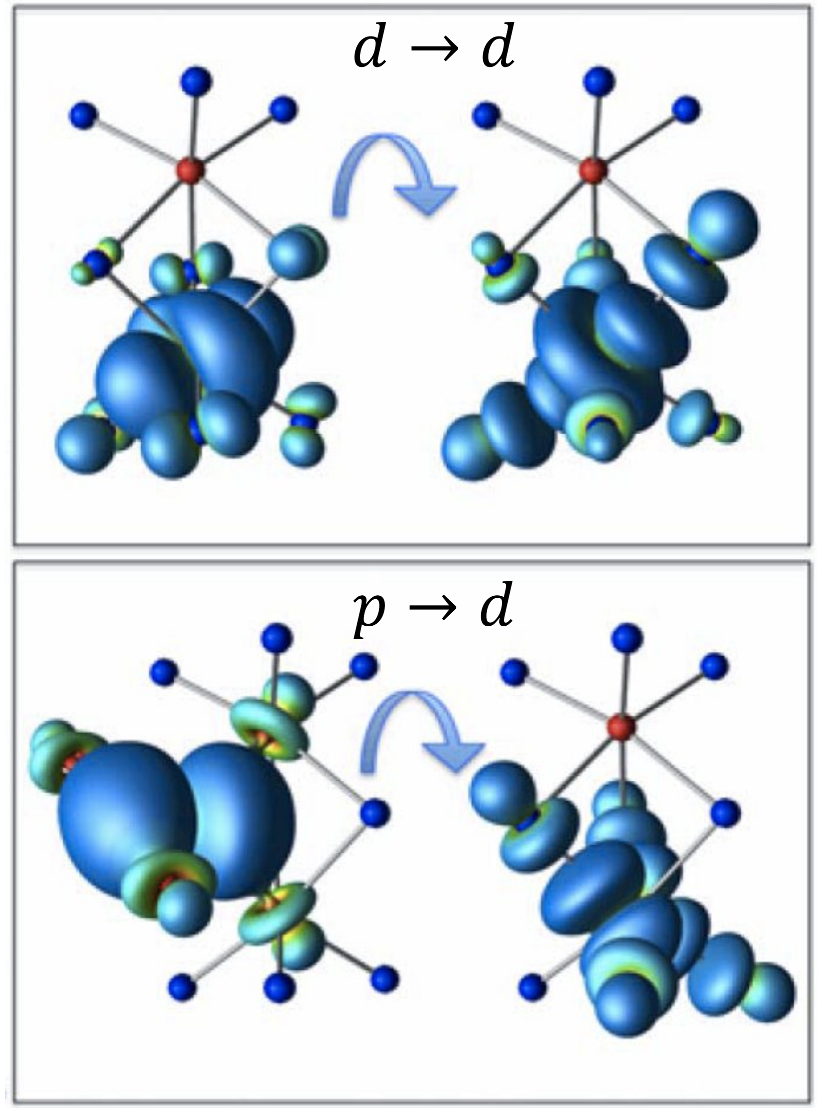
- V_2O_3 is one of the canonical model systems for a Mott-Hubbard insulator
- The Mott transition is usually explained within the Hubbard model with U = on-site Coulomb repulsion



Imada, Fujimori, and Tokura:
Metal-insulator transitions, RMP 70 1039 (1998)

V_2O_3

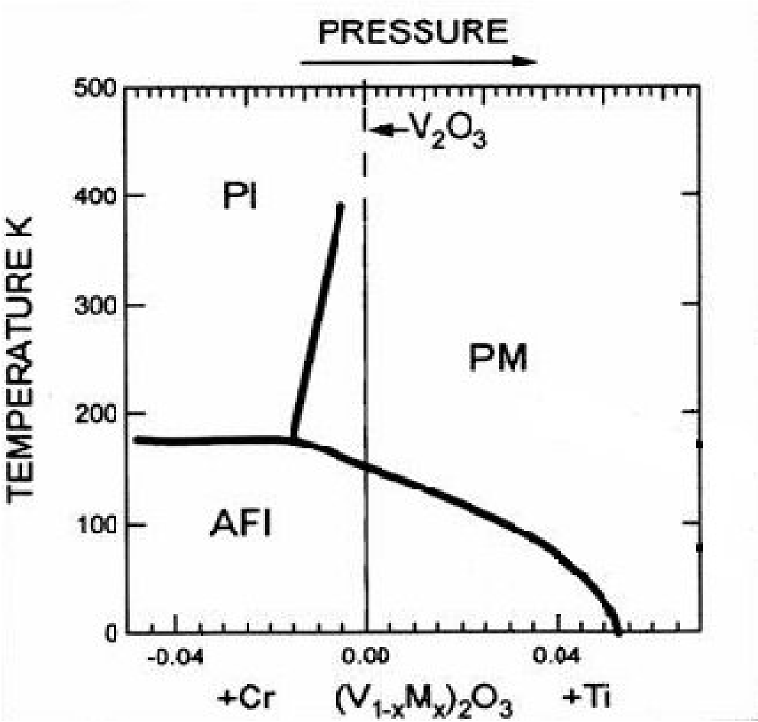




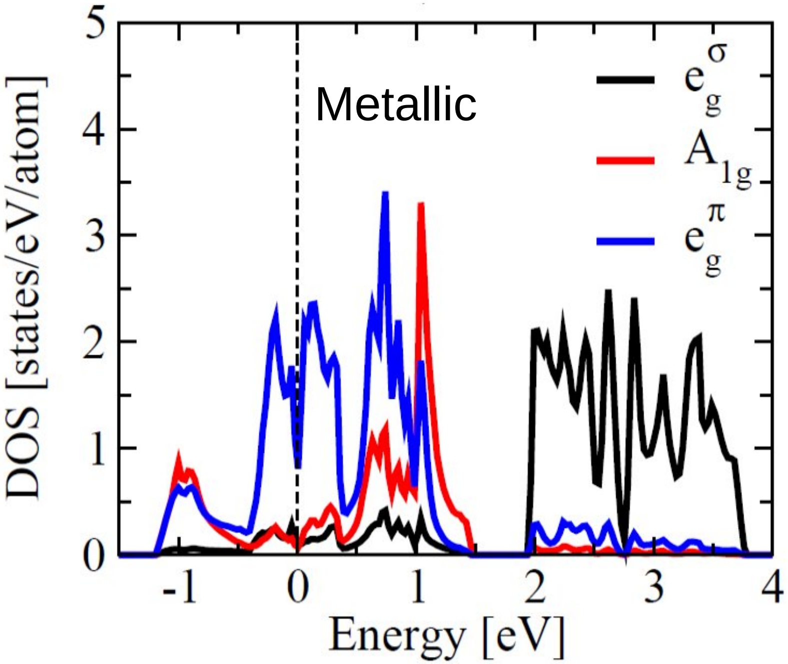
$$e^{i\mathbf{Q}\cdot\mathbf{r}} = 1 + i\mathbf{Q}\cdot\mathbf{r} - (\mathbf{Q}\cdot\mathbf{r})^2/2 + \cdots$$

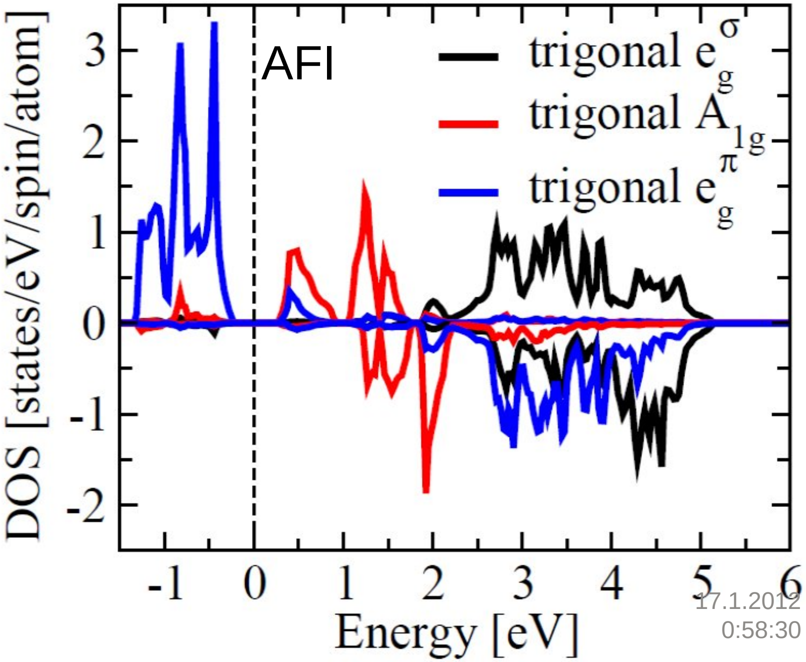
A.V.Kozhevnikov et al., in preparation

V_2O_3



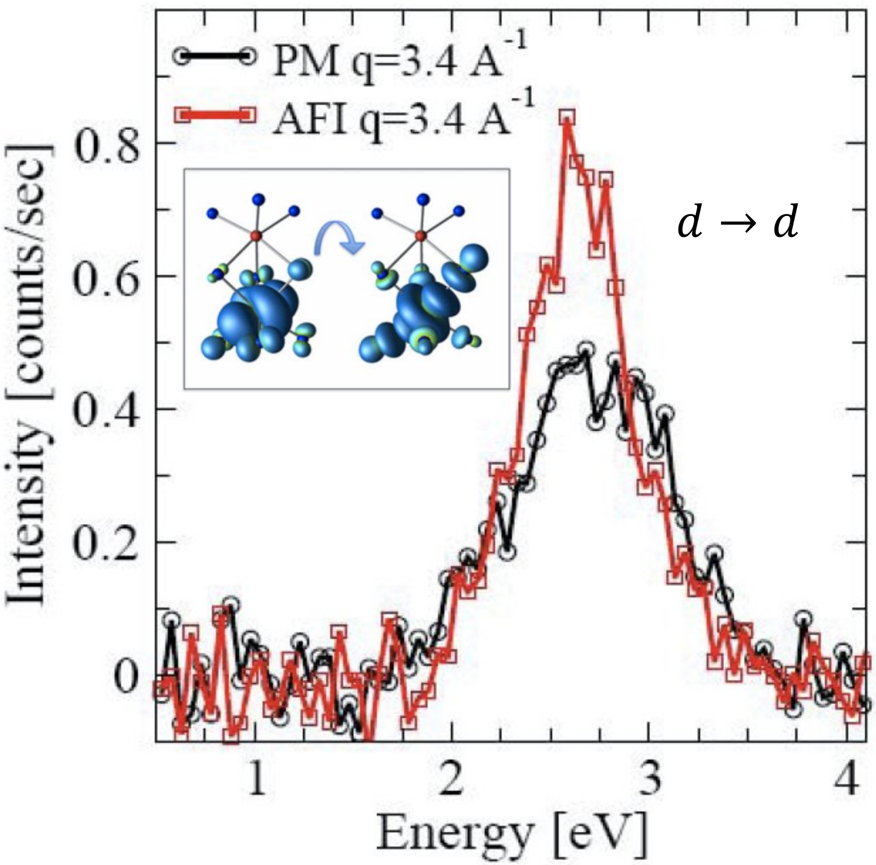
*McWhan et al., PRL **27** (1971)*

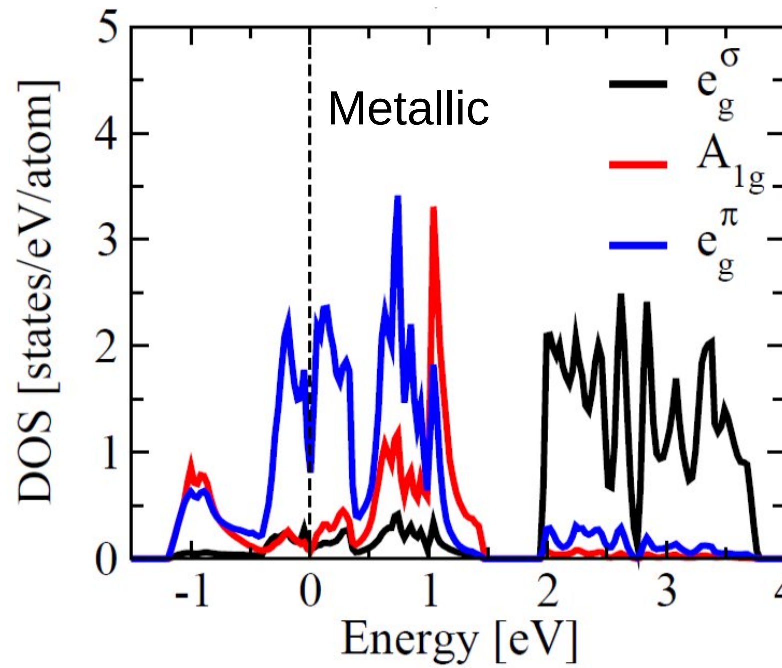


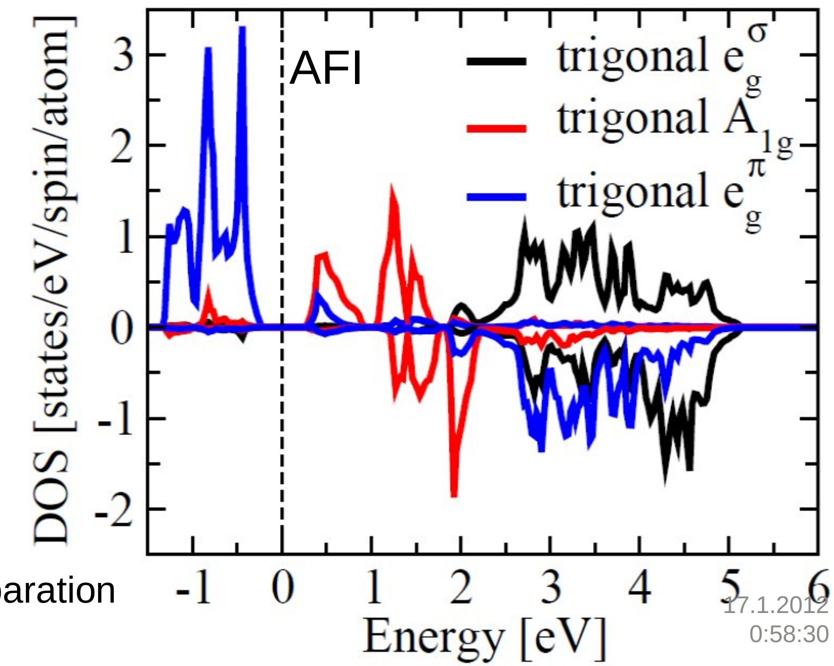


A.V.Kozhevnikov et al., in preparation

V_2O_3

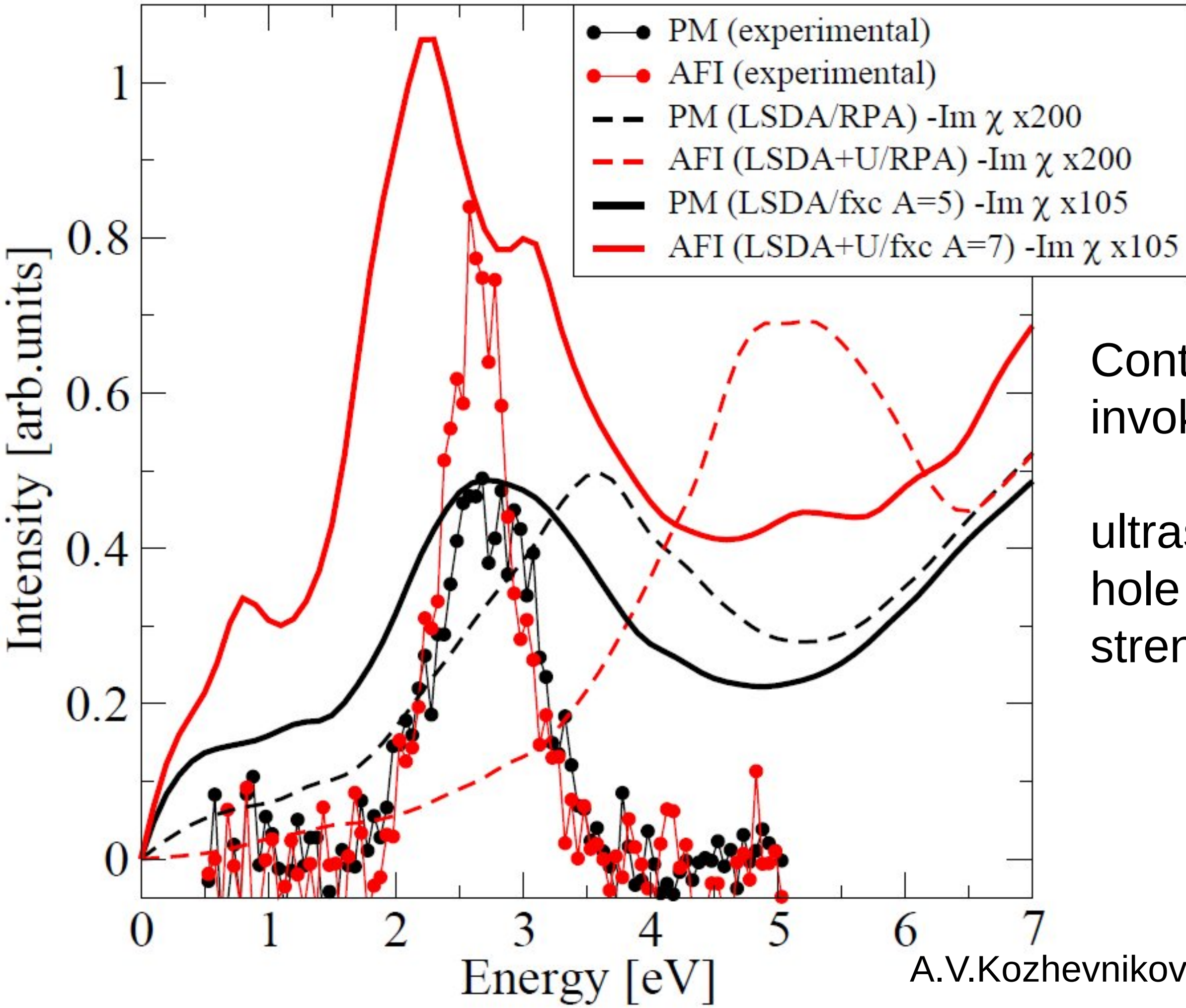






A.V.Kozhevnikov et al., in preparation

Metal-to-insulator transition in V_2O_3

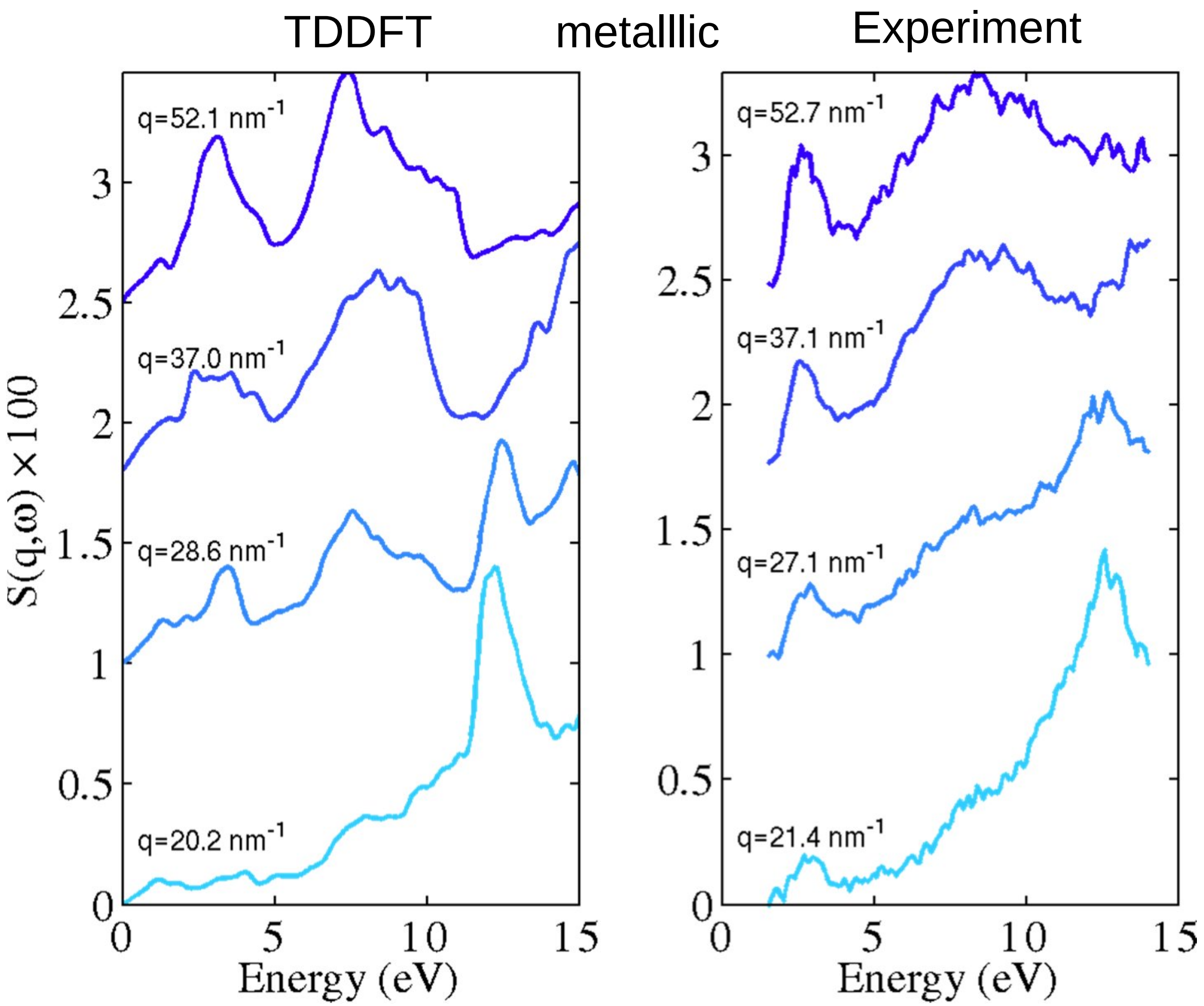


Contact exciton model invoked:

ultrashort-range electron-hole attraction, tunable strength with parameter A

A.V.Kozhevnikov et al., in preparation

V_2O_3



A.V.Kozhevnikov et al., in preparation

Metal-to-insulator transition in V_2O_3

Conclusions for metal-insulator transition of V_2O_3:

- d-d excitation changes surprisingly little upon going to insulating phase
- RPA would predict much larger change
- Must imply a switching-on of a excitonic interaction upon the MIT?

A.V. Kozhevnikov, M.C. Troparevsky, T. C. Schulthess, A.G. Eguiluz,
T. Pylkkänen, L. Simonelli, G. Monaco, and S. Huotari, under preparation

Outline

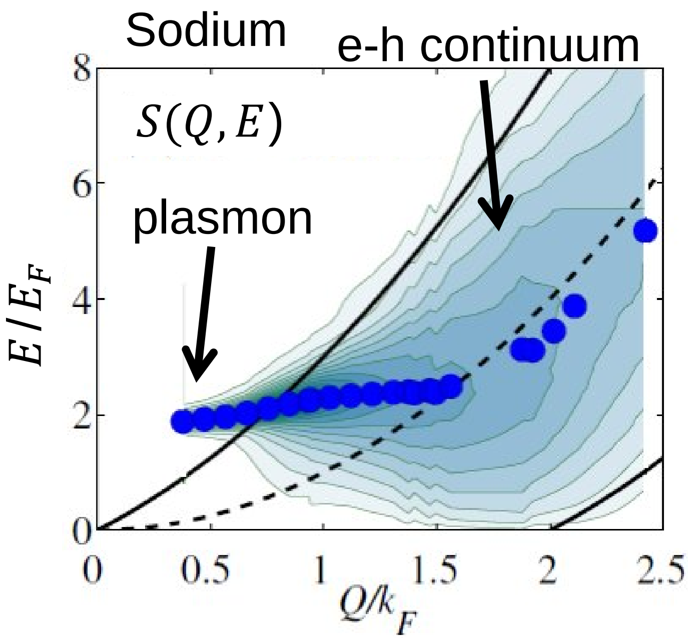
Part 1 Metal-to-insulator transition

Part 2 Plasmons

Part 3 Double plasmons

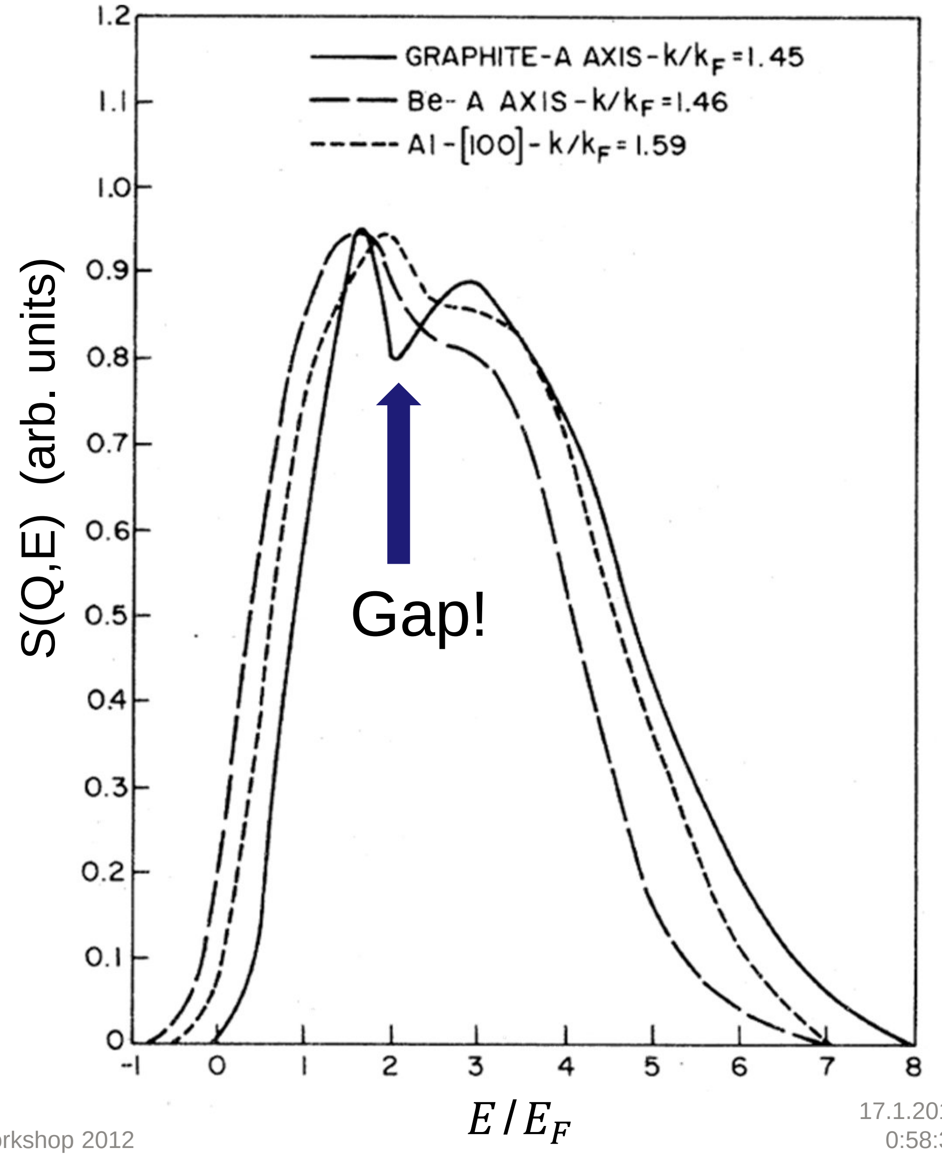
Part 4 Quasiparticle renormalisation

Electron gas response



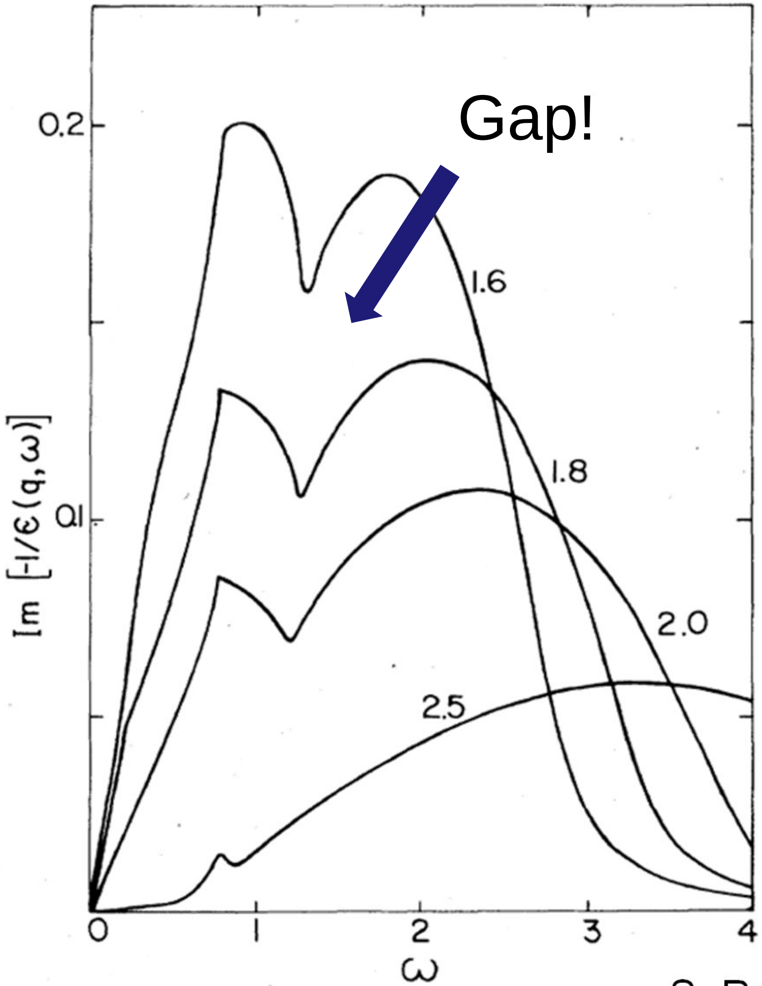
In 1974, $S(Q,E)$ for many materials seemed to suggest a ”universal shape” of the electron gas response function

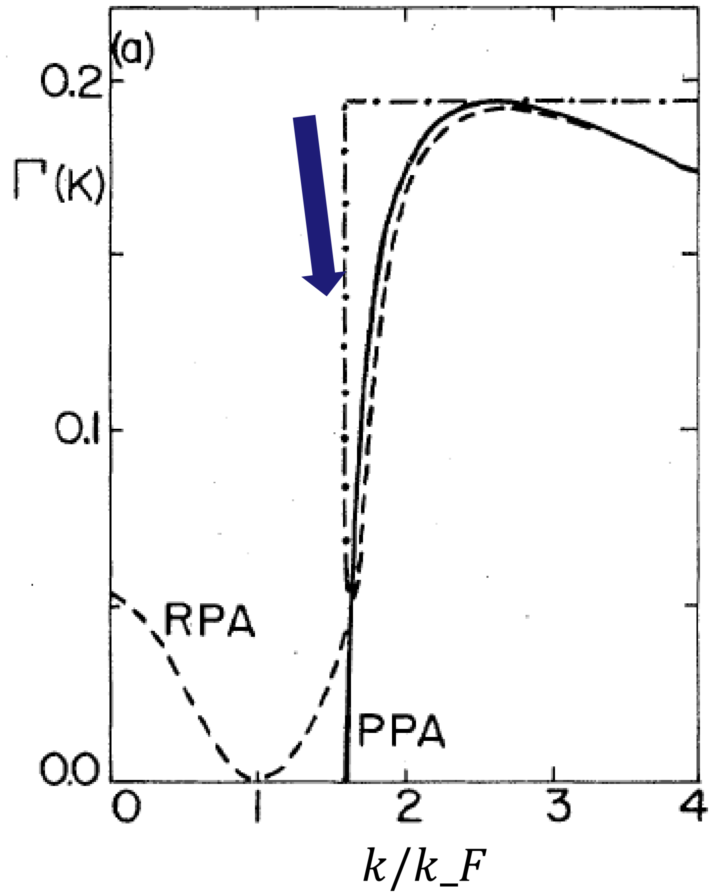
P.M.Platzman and P.Eisenberger, PRL 33, 152 (1974)



Theory for electron gas

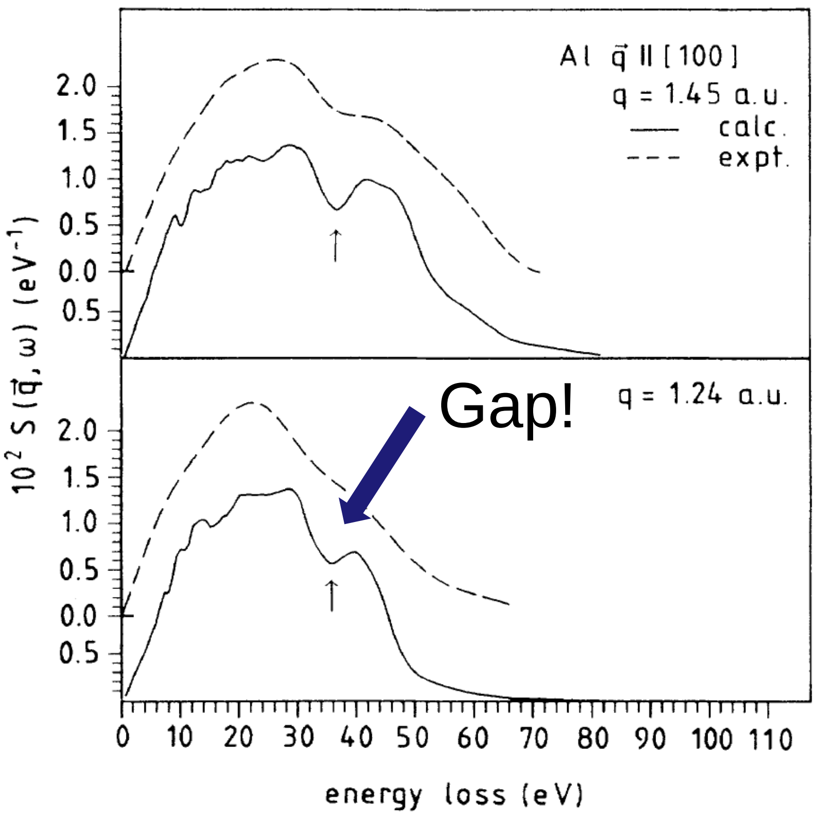
Gap structure appears when taking into account quasiparticle lifetimes





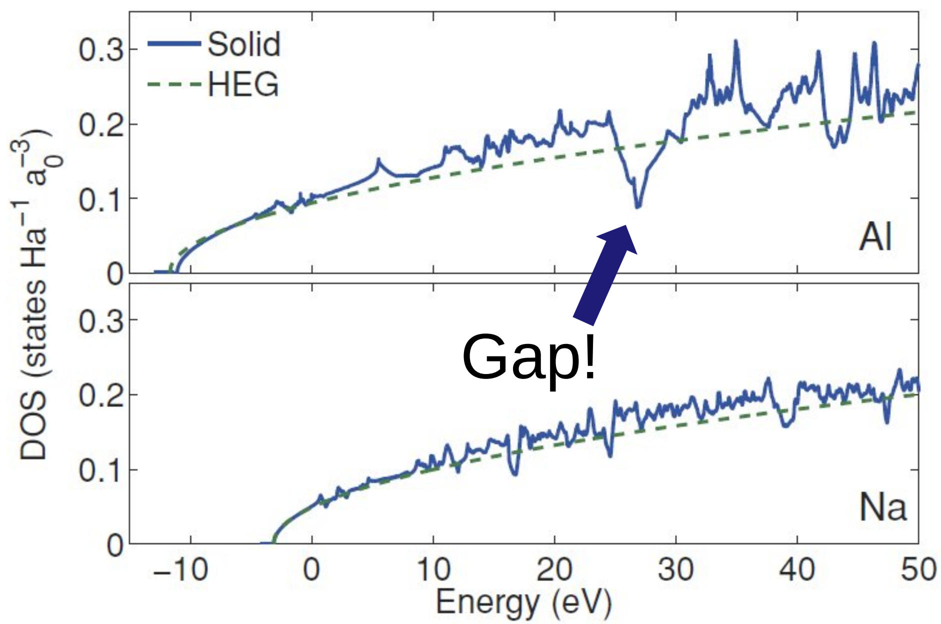
S. Rahman and G. Vignale, PRB 30, 6951 (1984)

Gap-structure in Al



Winfried Schülke et al. PRB 47, 12426 (1993)

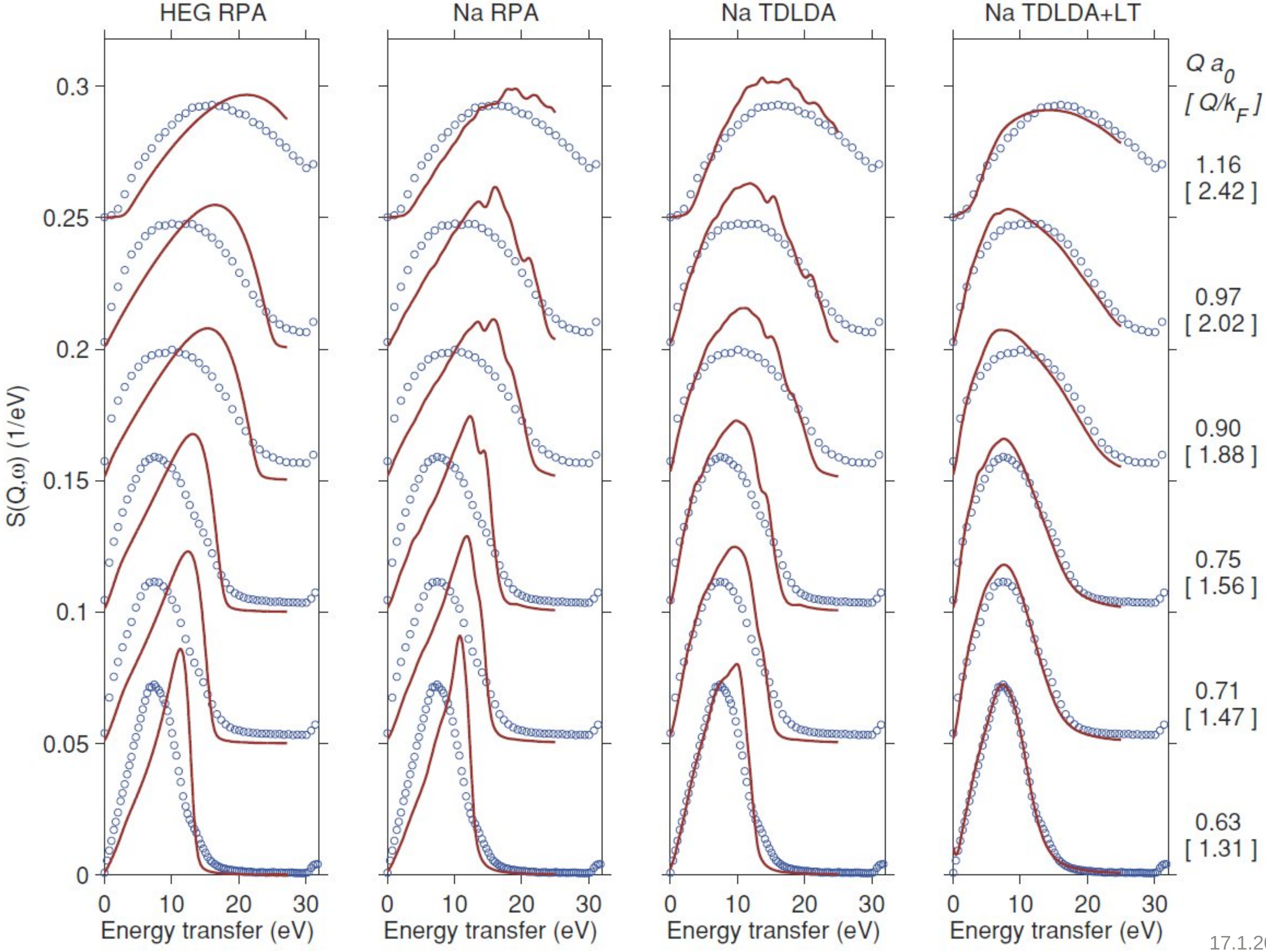
Explained to be a band-structure effect: a gap in the DOS



S. Huotari, M. Cazzaniga et al., PRB **84**, 075108 (2011)

Results for sodium

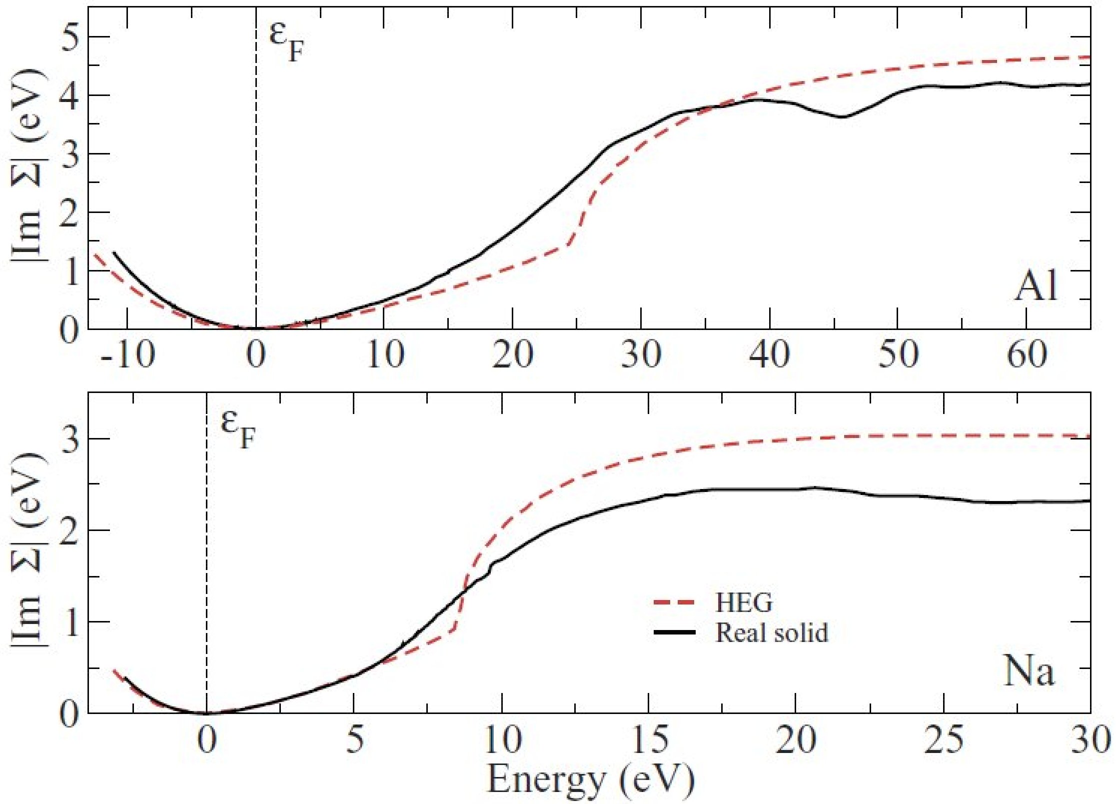
S. Huotari et al., PRB **84**, 075108 (2011)



TDLDA+LifeTimes (LT)

H.-C. Weissker et al., Phys. Rev. Lett. 97, 237602 (2006)
M. Cazzaniga et al., Phys. Rev. B 84, 075109 (2011)

$$\chi^0_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega) = -\frac{1}{V_{BZ}}\sum_{j,j'}\int_{BZ} d^3k[f(\epsilon_{j'}(\mathbf{k}+\mathbf{q})) - f(\epsilon_j(\mathbf{k}))]\frac{\langle \mathbf{k},j|e^{-i(\mathbf{q}+\mathbf{G})\cdot\hat{\mathbf{r}}}|\mathbf{k}+\mathbf{q},j'\rangle\langle \mathbf{k}+\mathbf{q},j'|e^{i(\mathbf{q}+\mathbf{G}')\cdot\hat{\mathbf{r}}}|\mathbf{k},j\rangle}{\omega - [\epsilon_{j'}(\mathbf{k}+\mathbf{q}) - \epsilon_j(\mathbf{k})] + i\eta}$$

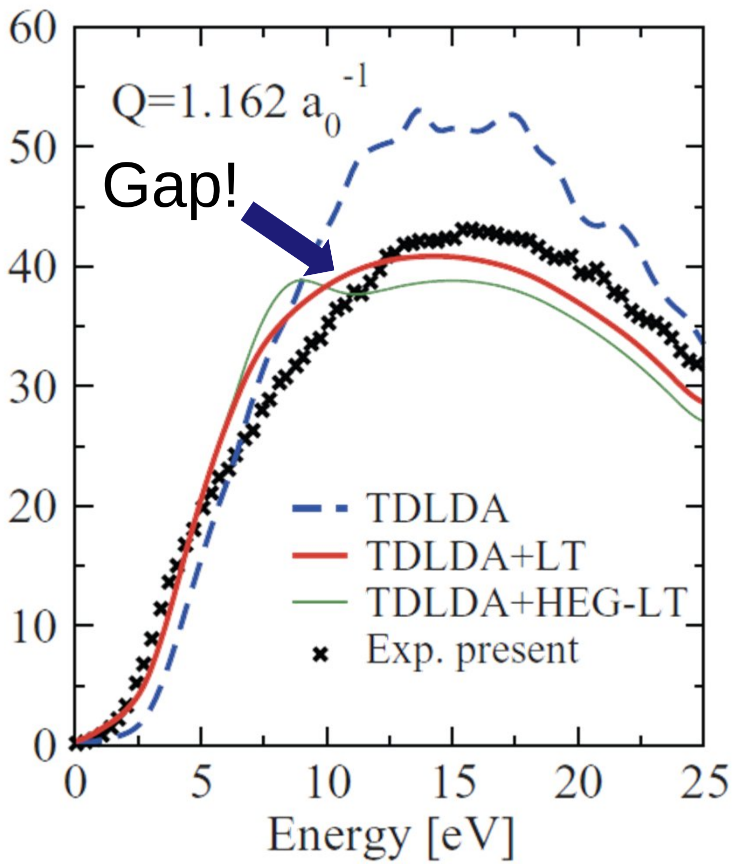


$$i|\mathrm{Im}\,\Sigma_{j'}(\mathbf{k}+\mathbf{q})| + i|\mathrm{Im}\,\Sigma_j(\mathbf{k})|$$

Inclusion of lifetimes via a modified independent particle polarizability χ_0^{LT}

Results for sodium

M. Cazzaniga et al., PRB **84**, 075109 (2011)



Using HEG lifetimes to HEG or real-metal Na, does a gap structure in the S(Q,E).

The gap is washed out if real-metal lifetimes are used.

Fictivious HEG does in fact keep the gap structure but Na is not close enough to the HEG!

Plasmons

Conclusions about plasmons in Na:

- Correlation effects beyond RPA surprisingly large in Na
- Quasiparticle lifetimes have to be taken into account properly
- Fine structure to the S(Q,E) expected in HEG; washed out in Na

PHYSICAL REVIEW B **84**, 075108 (2011)

Dynamical response function in sodium studied by inelastic x-ray scattering spectroscopy

Simo Huotari,[1,2,*] Marco Cazzaniga,[3,4] Hans-Christian Weissker,[4,5,6] Tuomas Pylkkänen,[1,2] Harald Müller,[2] Lucia Reining,[4,5] Giovanni Onida,[3,4] and Giulio Monaco[2]

PHYSICAL REVIEW B **84**, 075109 (2011)

Dynamical response function in sodium and aluminum from time-dependent density-functional theory

Marco Cazzaniga,[1,2,*] Hans-Christian Weissker,[2,3,4] Simo Huotari,[5,6] Tuomas Pylkkänen,[5,6] Paolo Salvestrini,[1,7] Giulio Monaco,[5] Giovanni Onida,[1,2] and Lucia Reining[2,3]

Outline

Part 1 Metal-to-insulator transition

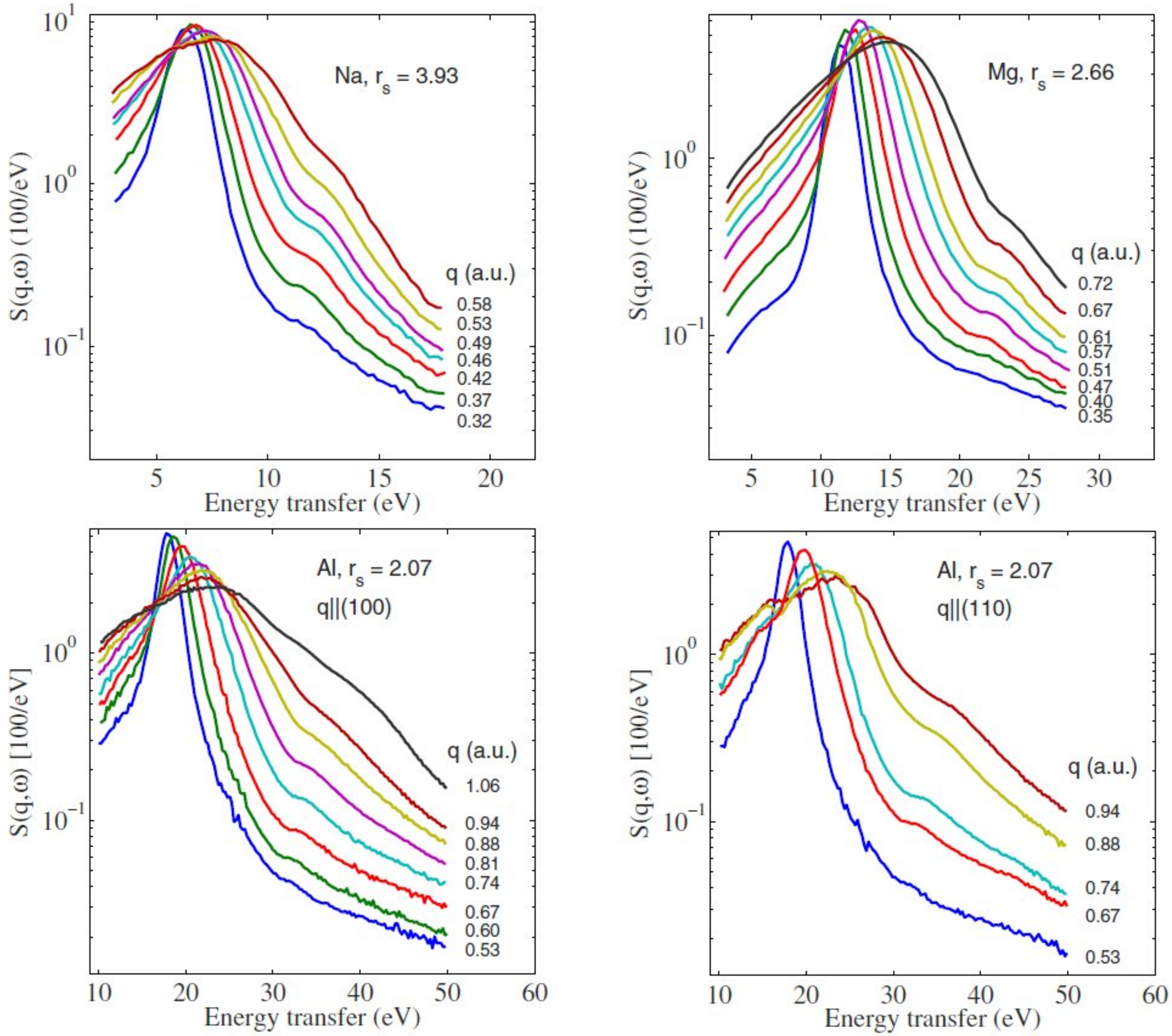
Part 2 Plasmons

Part 3 Double plasmons

Part 4 Quasiparticle renormalisation

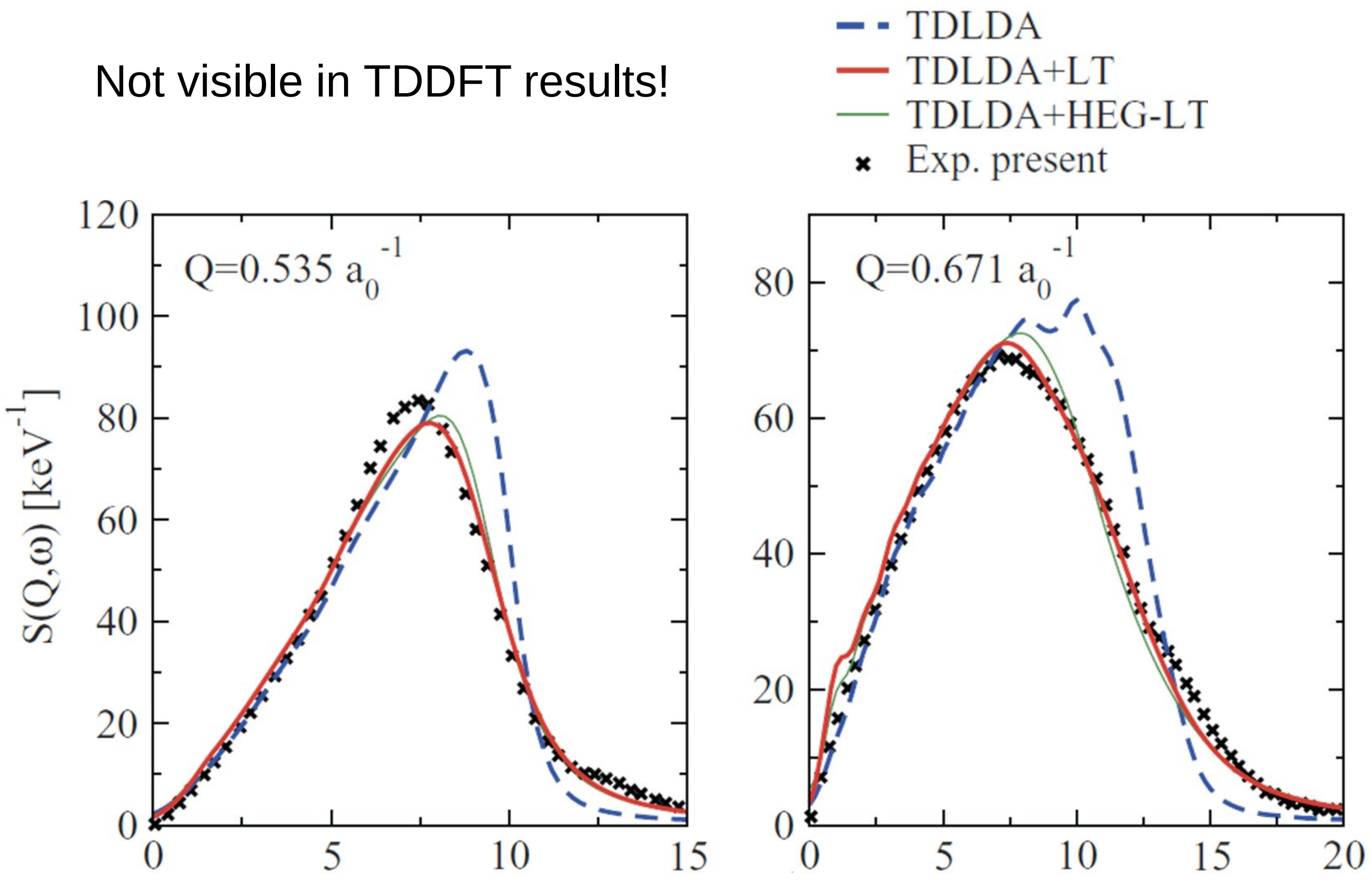
Double plasmons

S. Huotari et al.,
PRB **77**, 195125 (2008)



Double plasmons

Not visible in TDDFT results!



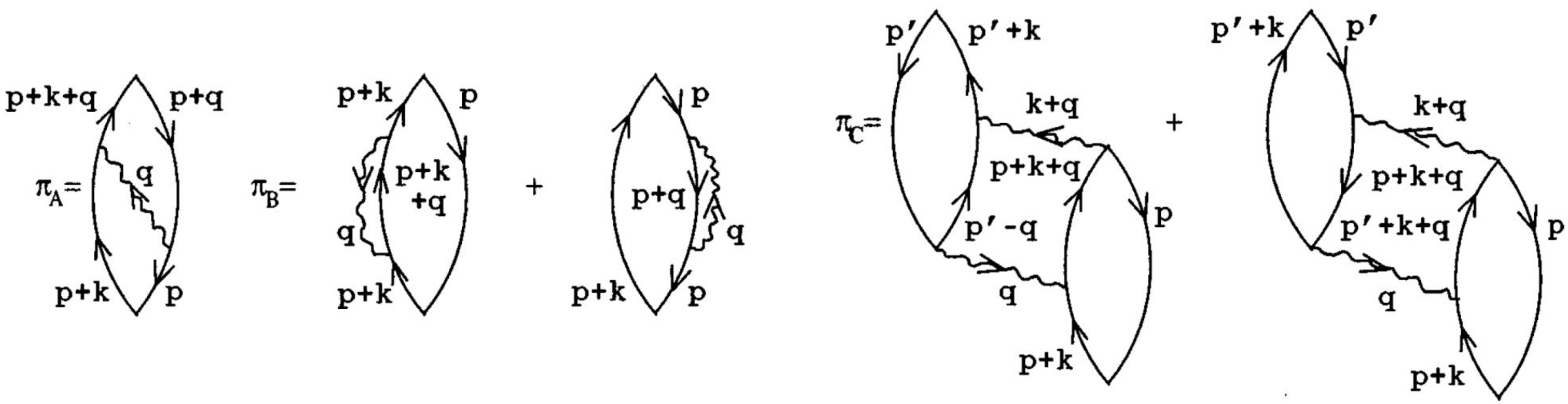
Energy transfer (eV)

M. Cazzaniga et al., PRB **84**, 075109 (2011)

Double plasmons

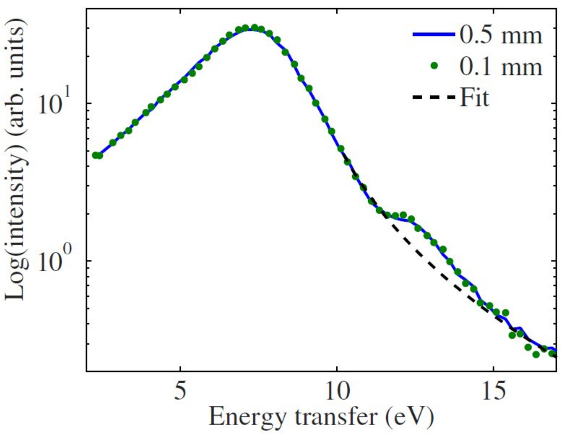
Need to go many-body!

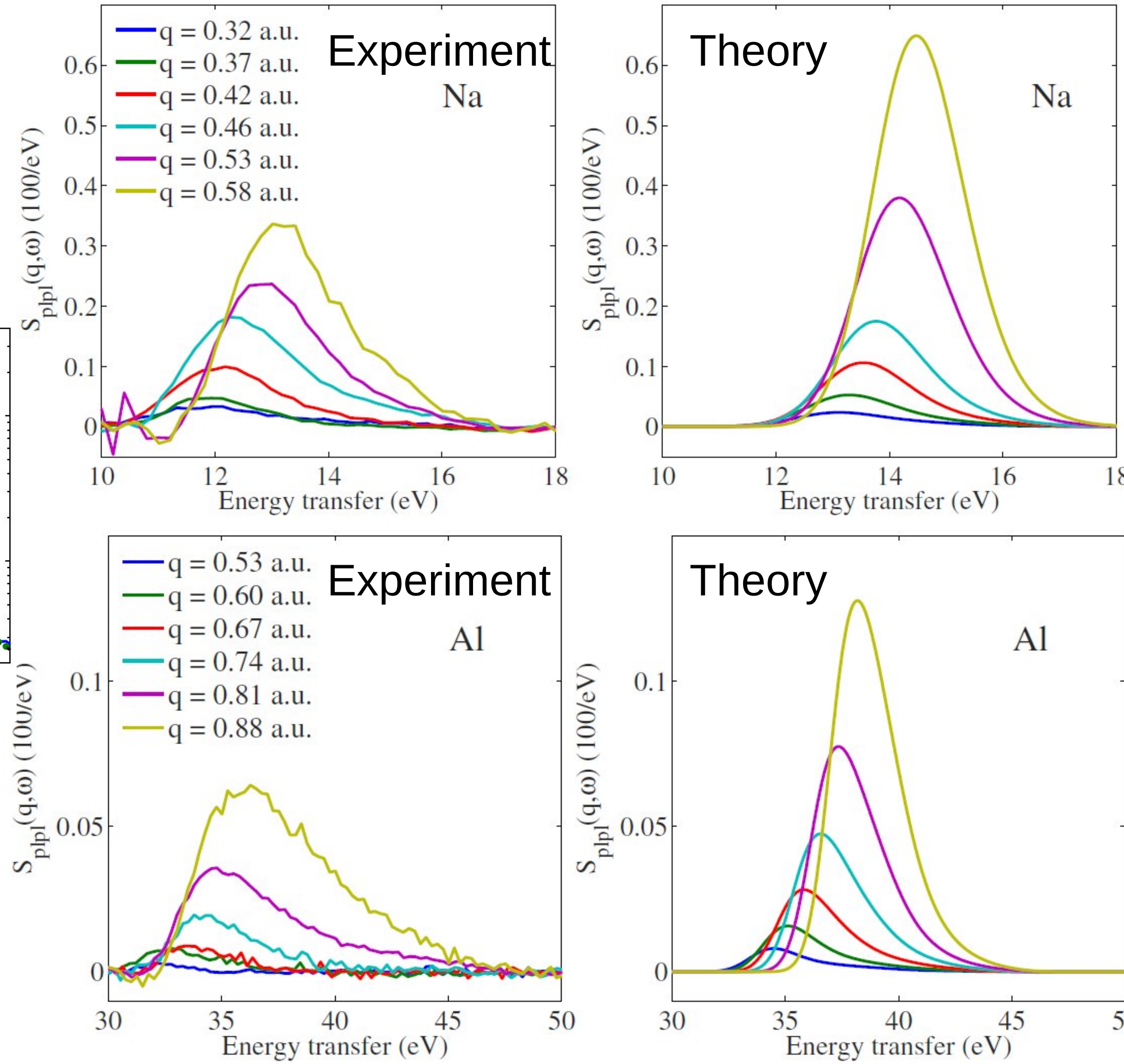
$$S(\mathbf{q},\omega) = \frac{q^2}{4\pi^2 n_0}\,\mathrm{Im}\left(\frac{-1}{\epsilon(\mathbf{q},\omega)}\right) \approx \frac{1}{\pi n_0} \times \frac{\mathrm{Im}[\pi_0(\mathbf{q},\omega) + \pi_\mathrm{A}(\mathbf{q},\omega) + \pi_\mathrm{B}(\mathbf{q},\omega) + \pi_\mathrm{C}(\mathbf{q},\omega)]}{\epsilon_\mathrm{L}^2(\mathbf{q},\omega)}$$



K. Sturm and A. Gusarov, PRB 62, 16474 (2000)

Double plasmons





S. Huotari et al.,
PRB **77**, 195125 (2008)

Double plasmons

Conclusions about double plasmons in Al,Na and Mg:

- Double plasmons observed in S(Q,E), not explained by TDDFT
- Many-body effects explain the spectra to ~2 eV and factor of 2 in intensity
- Should one try to incorporate the appropriate interaction in f_{xc} in TDDFT?

PRL **95,** 157401 (2005) PHYSICAL REVIEW LETTERS week ending 7 OCTOBER 2005

Correlation-Induced Double-Plasmon Excitation in Simple Metals Studied by Inelastic X-Ray Scattering

C. Sternemann,[1] S. Huotari,[2] G. Vankó,[2] M. Volmer,[1] G. Monaco,[2] A. Gusarov,[3,4] H. Lustfeld,[3] K. Sturm,[3] and W. Schülke[1]

PHYSICAL REVIEW B **77**, 195125 (2008)

Electron-density dependence of double-plasmon excitations in simple metals

S. Huotari,[1] C. Sternemann,[2] W. Schülke,[2] K. Sturm,[3] H. Lustfeld,[3] H. Sternemann,[2] M. Volmer,[2] A. Gusarov,[4] H. Müller,[1] and G. Monaco[1]

Outline

Part 1   Metal-to-insulator transition

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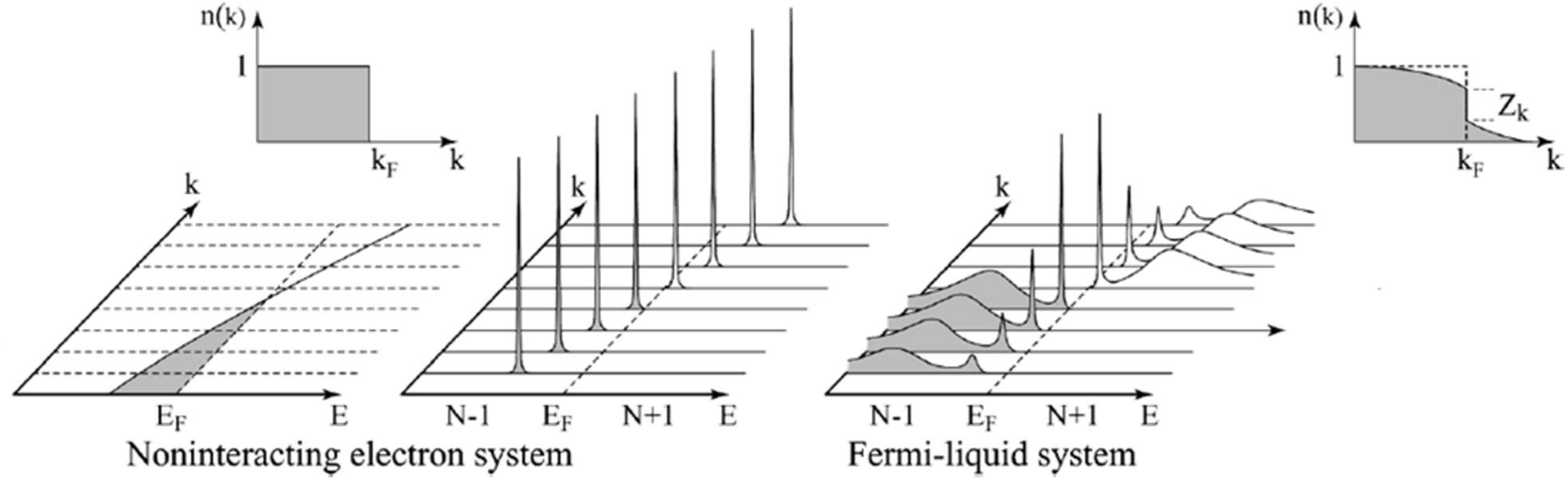
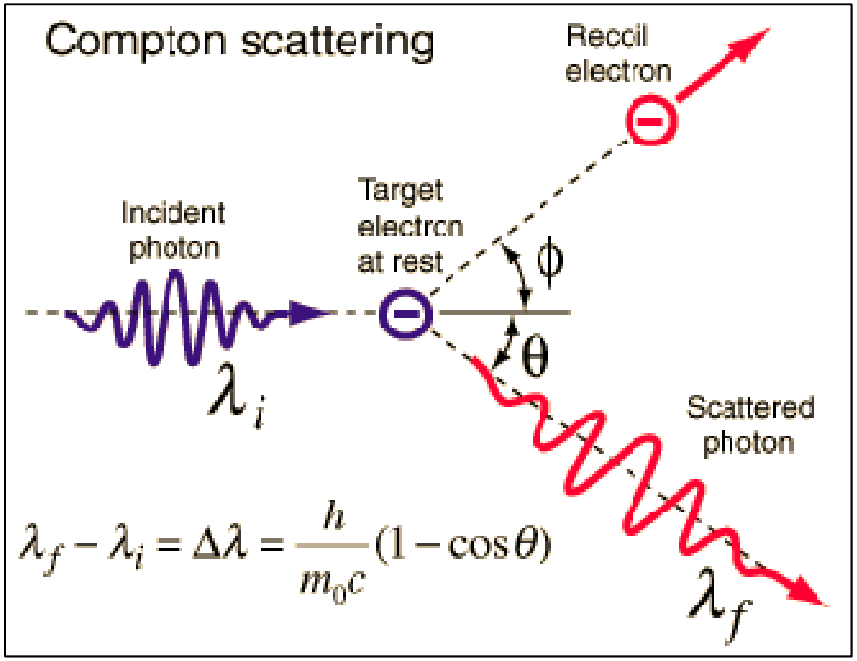
Quasiparticle renormalisation



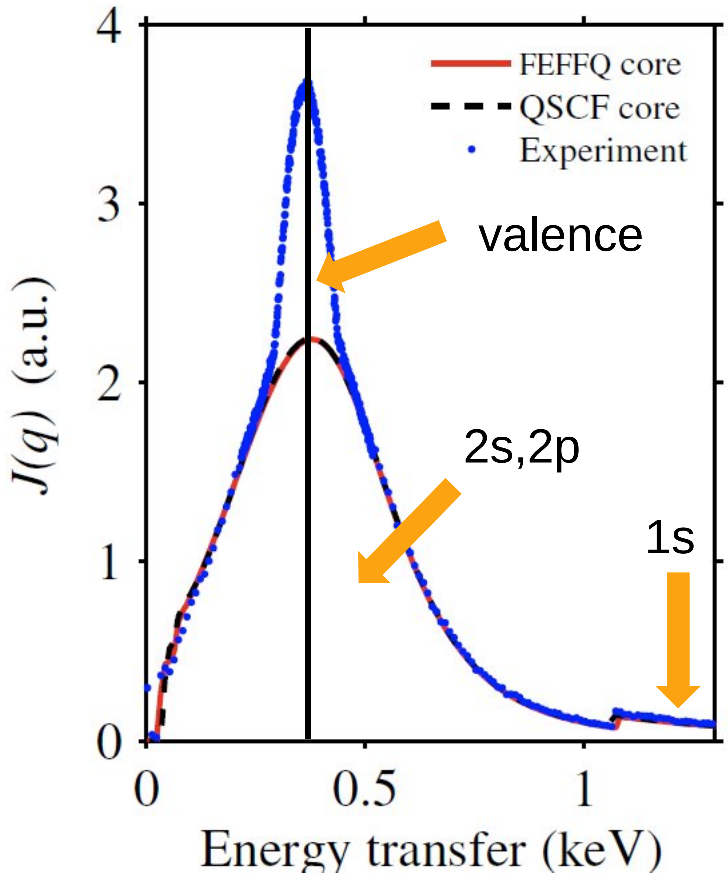
$$\mathrm{PES} \propto \sum_{f,i} \left| M^{\mathbf{p}}_{f,i} \right|^2 A(\mathbf{p}, E)\, \delta(E_K + E_m^{N-1} - E_i^N - \omega_1)$$

A. Damascelli et al.,
Rev. Mod. Phys 75, 473 (2003)

Compton spectroscopy



http://hyperphysics.phy-astr.gsu.edu/hbase/quantum/compton.html

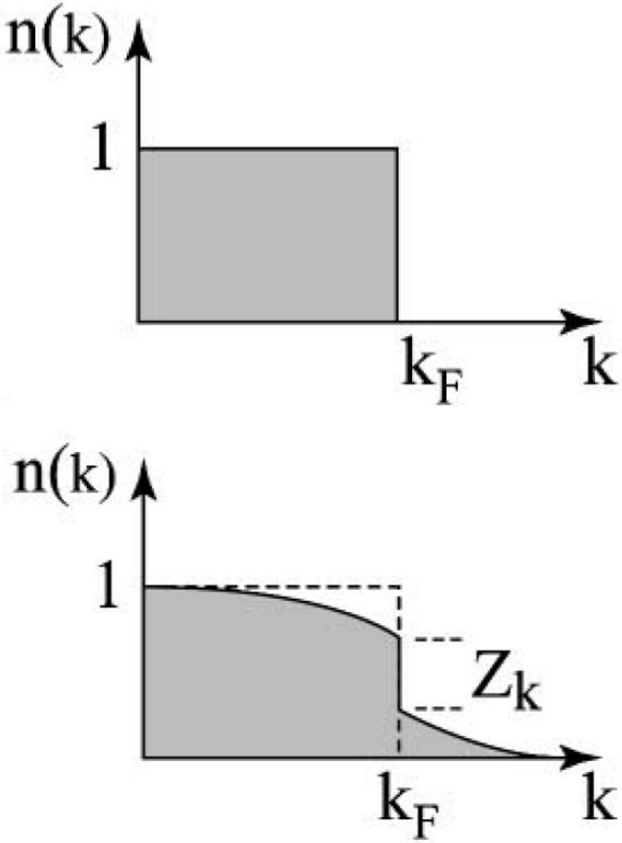


Compton profile $J(q)$

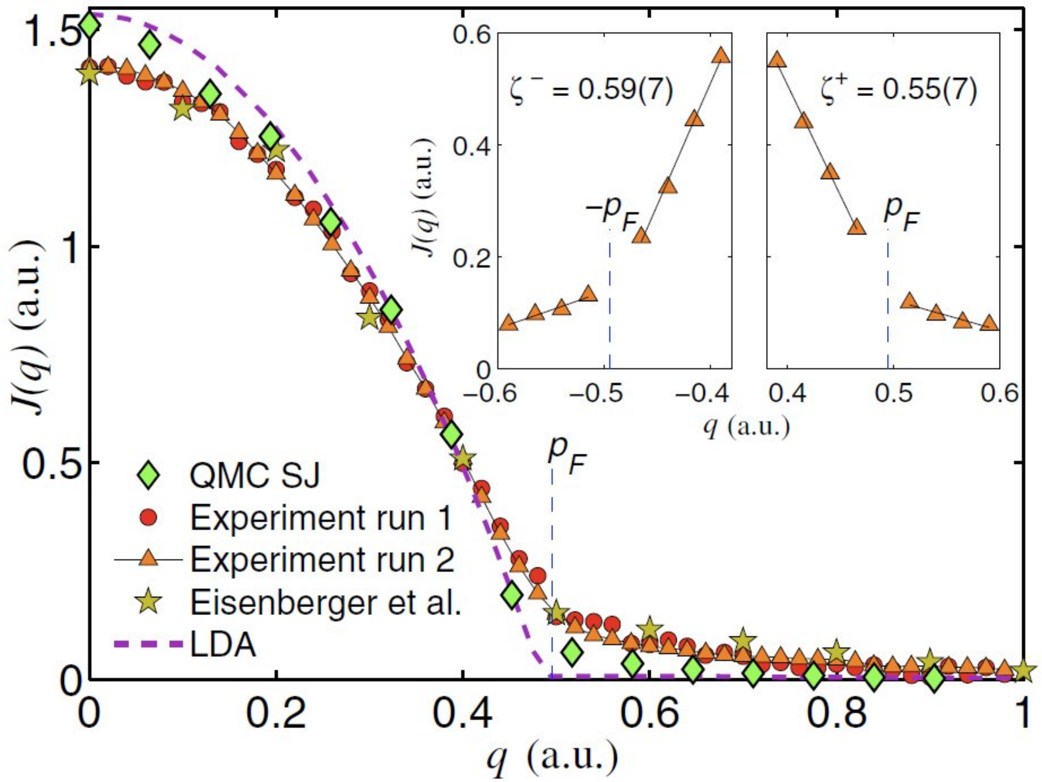
$$J(q) = \frac{3}{8\pi p_F^3} \int_{4\pi} d\Omega \int_{|q|}^{\infty} p n(\mathbf{p}) dp$$

S. Huotari et al.,
PRL 105, 086403 (2010)

Compton spectroscopy



Independent electrons:
$J(q) \propto (p_F^2 - q^2)$ for $|q| < p_F$



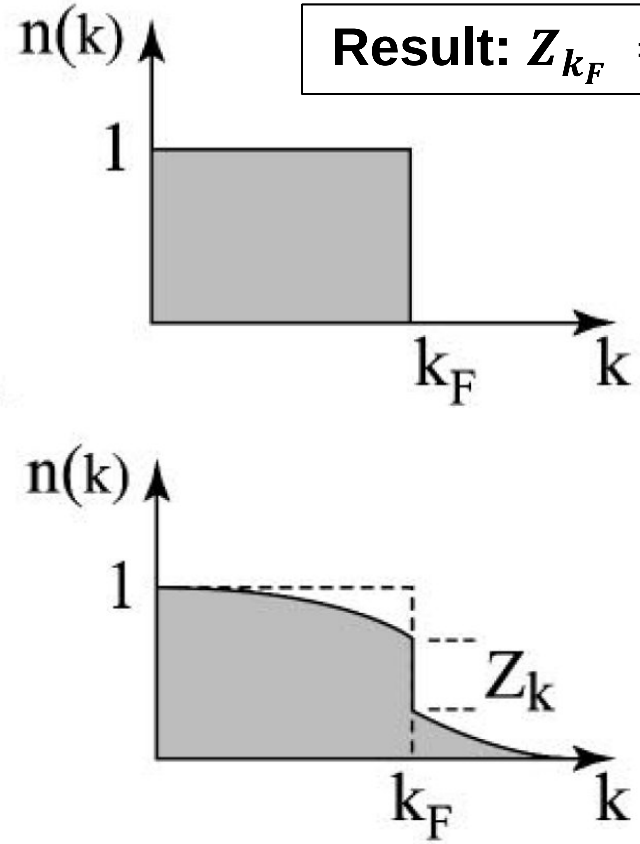
A. Damascelli et al.,
Rev. Mod. Phys 75, 473 (2003)

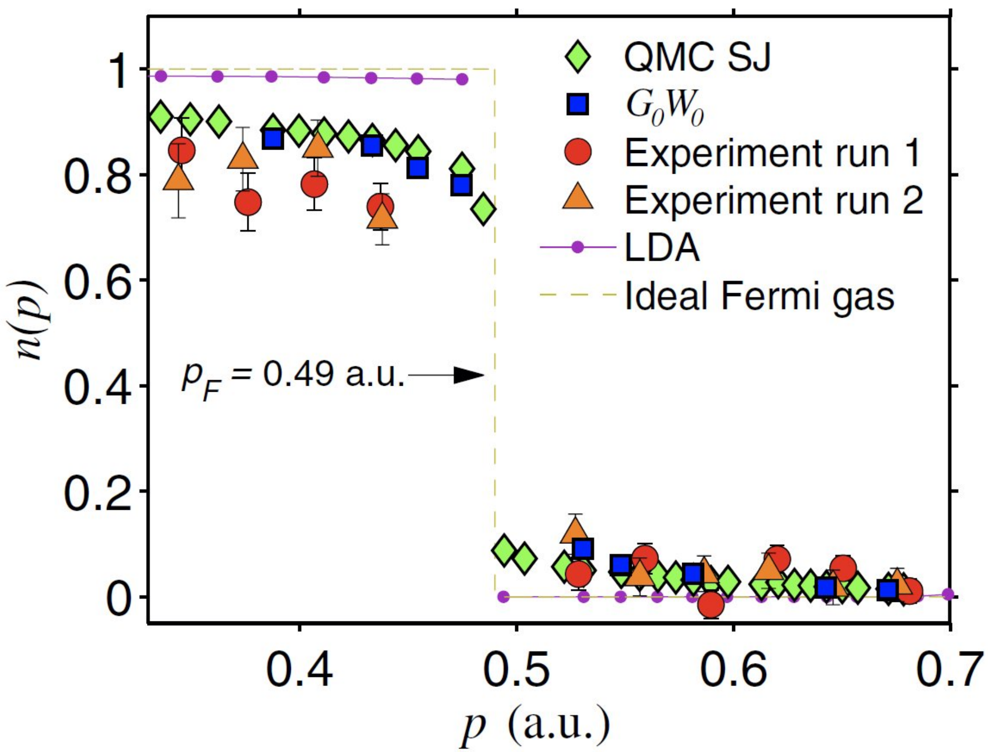
S. Huotari et al.,
PRL 105, 086403 (2010)

Momentum density of the electron gas

Result: $Z_{k_F} = 0.58 \pm 0.07$ for Na

$$n(p) = -\frac{2p_F^3}{3p}\frac{dJ(q)}{dq}\Big|_{q=p}$$





S. Huotari et al., PRL 105, 086403 (2010)

Quasiparticle renormalisation

Conclusions about the QP renormalisation factor:

- Is in principle the area under the coherent peak in photoemission
- Can be independently measured by momentum-density experiment
- Good results and agreement with theory obtained for Na

PRL **105,** 086403 (2010) PHYSICAL REVIEW LETTERS week ending 20 AUGUST 2010

Momentum Distribution and Renormalization Factor in Sodium and the Electron Gas

Simo Huotari,[1,2] J. Aleksi Soininen,[2] Tuomas Pylkkänen,[1,2] Keijo Hämäläinen,[2] Arezki Issolah,[3] Andrey Titov,[4] Jeremy McMinis,[5] Jeongnim Kim,[5] Ken Esler,[5] David M. Ceperley,[5] Markus Holzmann,[6] and Valerio Olevano[4]

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[5]*Department of Physics and NCSA, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801, USA*
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A. Eguiluz, A.V.Kozhevnikov, M.C.Troparevsky, T.C.Schulthess et al.