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Consistency in atomic-scale calculations for strongly coupled systems

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Summary: Consistency is a key component of reliable predictions in strongly coupled systems

- Bulk thermal and electrical transport:
  - Critical for hydrodynamic simulations
  - Widely used Drude & Ziman formulations have multiple components
  - Errors can arise if these are not all computed in the same basis

- X-ray Thomson Scattering:
  - Has become a popular diagnostic for warm dense matter
  - Widely used models take a piecemeal approach to Chihara separation

- X-ray emission and absorption from dense plasmas:
  - Important for both hydrodynamics and diagnostics
  - Models tend to be either detailed with *ad-hoc* plasma effects or rough but consistent; neither appears adequate for, *e.g.*, recent LCLS data

Opinion: internal consistency should be emphasized in model development and comparisons with experimental data performed wherever possible.

Transport: Ziman and Drude approaches have multiple components

Extended Ziman

Drude

$$\eta = -\frac{\Omega}{3\pi}\frac{1}{Z_0 Z_i}\int_0^{\infty}\frac{\partial f(\varepsilon;\mu,T)}{\partial\varepsilon}\left\{\int_0^{2p} k^3\left(\frac{d\sigma(p,\theta)}{d\theta}\right)S(k)\,dk\right\}d\varepsilon = \frac{\Omega}{Z_i}\left(\frac{1}{\tau_{ei}}\right)$$

Electronic components:

Z_i is the (ill-defined*) number of conduction electrons

$f(\varepsilon)$ is the Fermi function, dependent on μ

$\frac{d\sigma(p,\theta)}{d\theta}$ is the differential scattering cross section

Ionic component:

S(k) is the static ion-ion structure factor, which plays a major role in strongly coupled systems

In principle (and sometimes in practice**), these components can be taken from independent sources.

*Murillo et al., Phys Rev. E 87, 063113 (2013)

**Hansen et al., UCRL-PROC-218150 (2005)

Measurements of Drude components indicate that compensating errors can give false agreement

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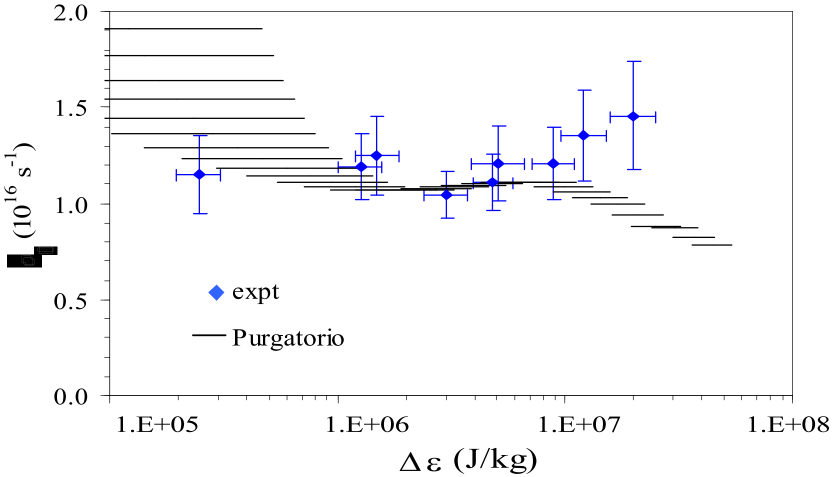
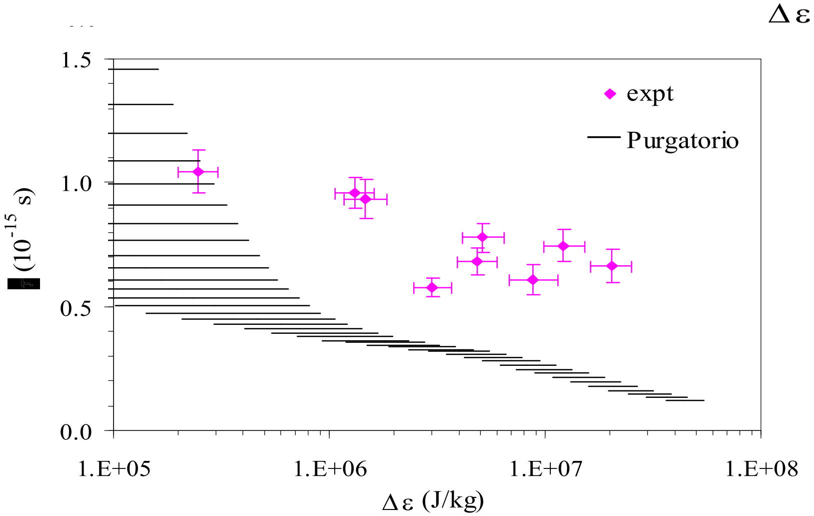
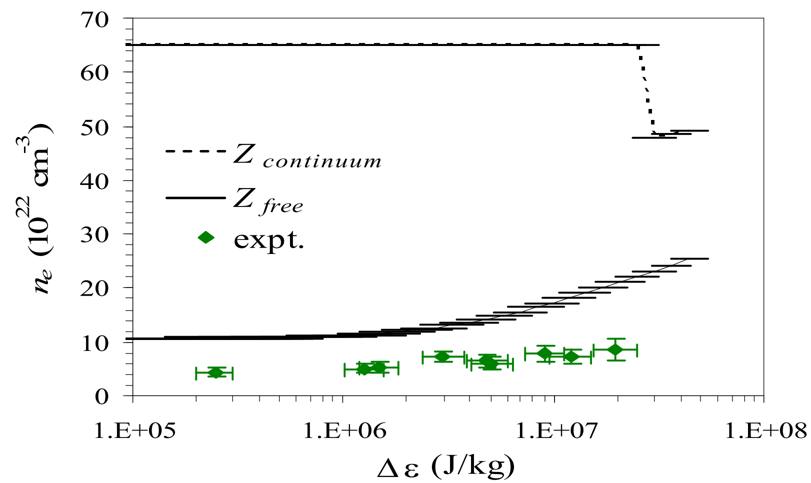
$$\eta = \frac{\Omega}{Z_i}\left(\frac{1}{\tau_{ei}}\right)$$



Apparently reasonable agreement between Purgatorio conductivities and experiments* was belied by detailed measurements

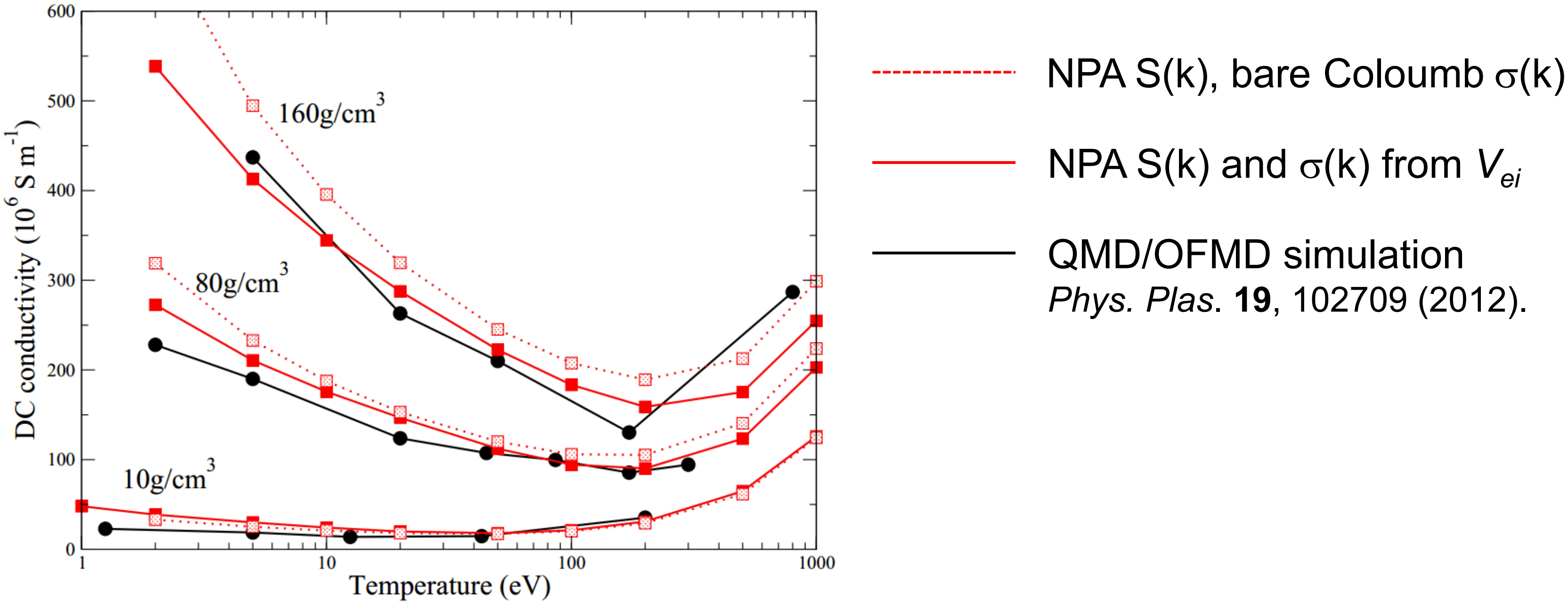




A consistent model at least has the advantage of accounting for the interplay between the components (*e.g.* increasing v_{xc} can increase τ_{ei} and decrease Z_i)

Widmann et al., PRL 92 125002 (2004) and Ao et al., PRL 96 055001 (2006)

Consistency between S(k) and σ(k) via the electron-ion potential also appears important



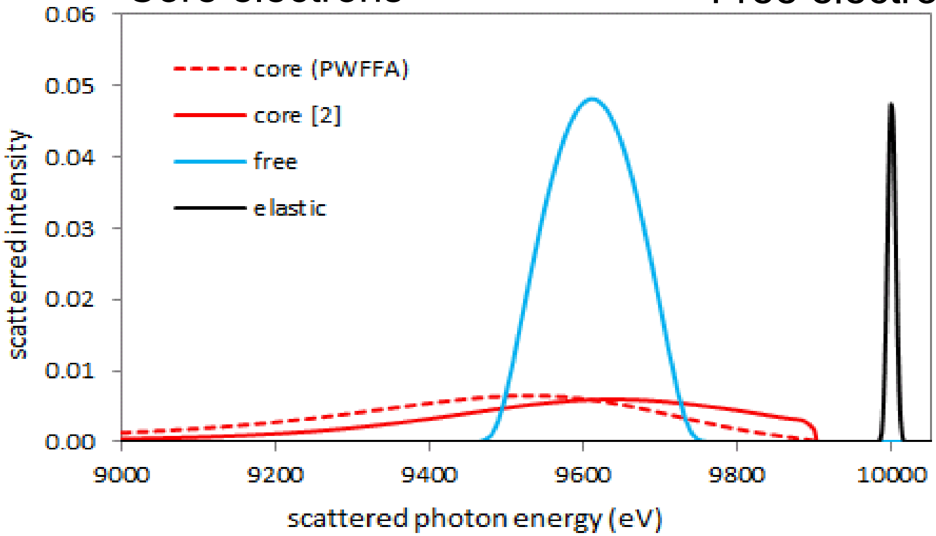
The Neutral Pseudo-Atom models of Starrett & Saumon[1], Perrot & Dharma-Wardana[2], and Faussurier[3] provide everything required for consistency.

1. C. Starrett and D. Saumon, High Energy Density Phys. **10**, 35 (2014).
2. F. Perrot and M.W.C. Dharma-Wardana, Phys. Rev. E **52**, 5352 (1995).
3. G. Faussurier, C. Blancard, P. Cossé, and P. Renaudin, Phys. Plasmas **17**, 052707 (2010).

XRTS: The Chihara approach splits the x-ray Thomson scattering signal into three components

Note dependence on ill-defined Z*

$$S_{ee}^{tot}(k,\omega) = \underbrace{Z_c \int d\omega' S_{ce}(\omega - \omega') S_s(k,\omega')}_{\text{Core electrons}} + \underbrace{Z_f S_{ee}^{(0)}(k,\omega)}_{\text{Free electrons}} + \underbrace{|n_f(k) + n_c(k)|^2 S_{ii}(k,\omega)}_{\text{Elastic component}}$$



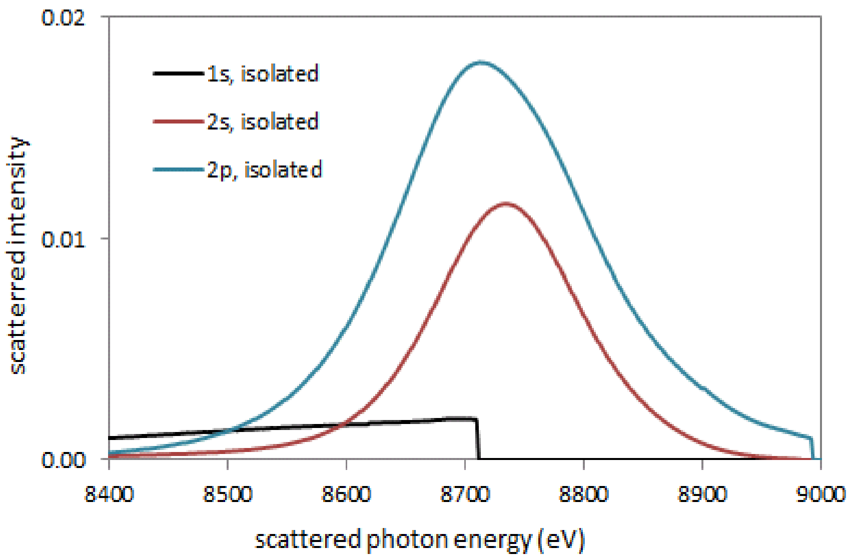
Widely used models treat each of these components independently and parameterize strongly coupled systems with Z*.

The NPA model presented in Souza *et al.*,[1] (*cf.* W.R. Johnson[2] and B. Mattern & G. Seidler[3]) appears to provide the most consistent treatment so far.

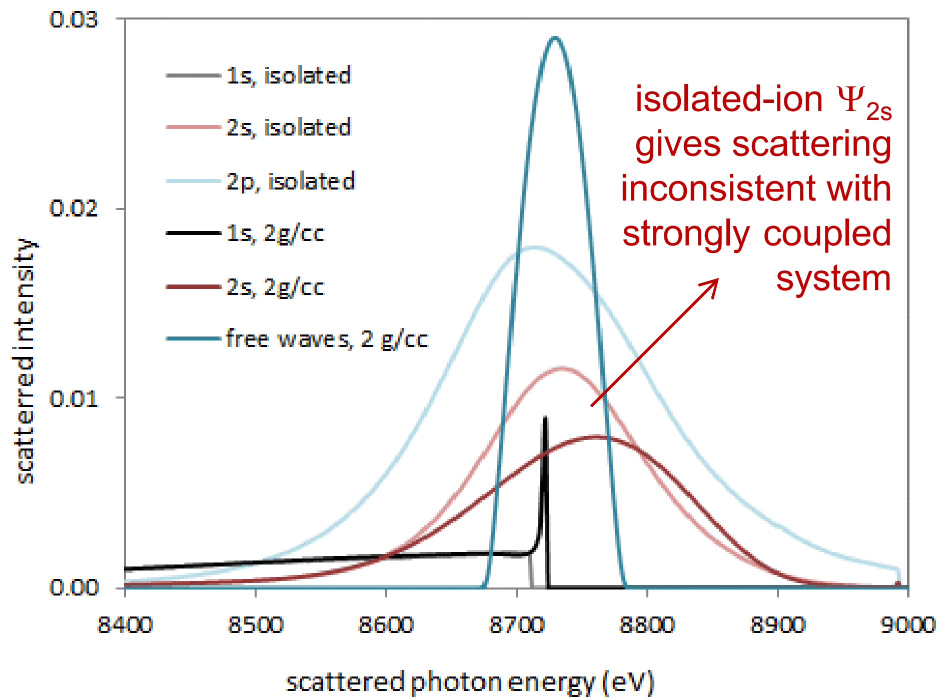
1. A. Souza, D. Perkins, C. Starrett, D. Saumon, and S. Hansen. Phys Rev. E **89**, 023108 (2014).
2. W.R. Johnson, J. Nilsen, and K.T. Cheng, Phys. Rev. E **86**, 036410 (2012).
3. B.A. Mattern and G.T. Seidler, Phys. Plasmas **20**, 022706 (2013).

What happens to the scattering signal as bound electrons become pressure ionized?

135° scattering from cold, isolated carbon:
Valence 2p state is bound



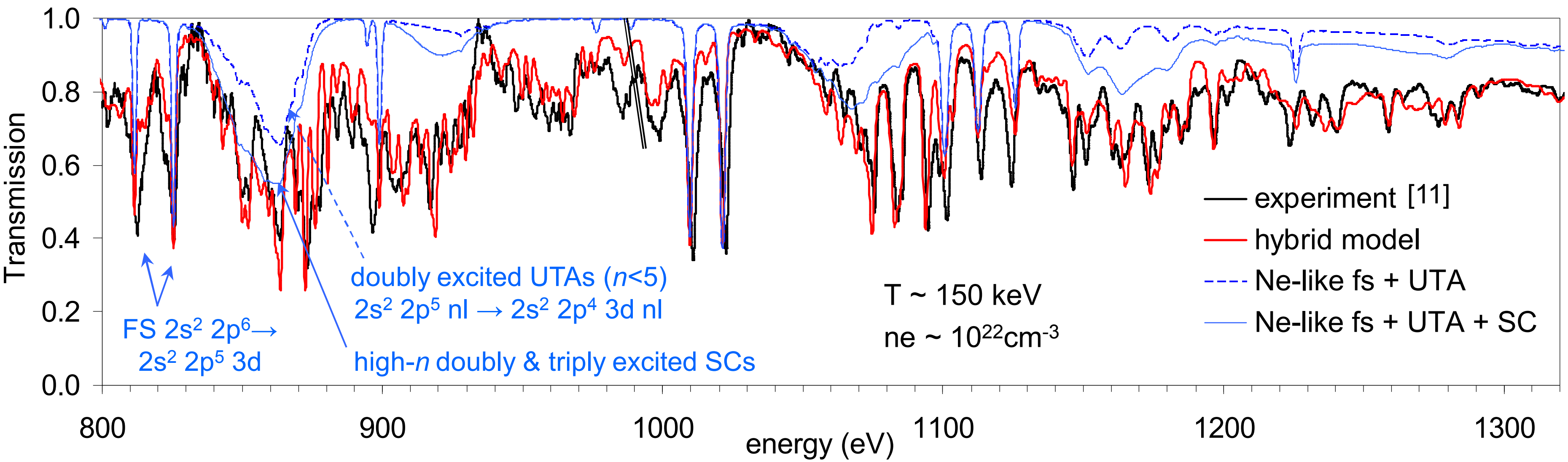
Scattering from cold carbon at 2 g/cc:
2p is pressure ionized (but not to a plane wave!)
1s and 2s scattering signals are modified



See poster of L. Johnson investigating the effect of continuum-wave distortion on free-free scattering signals.

X-ray emission and absorption models require extensive detail, completeness, and consistency

X-ray spectra can be measured with exquisite accuracy, revealing highly detailed electronic structure "supported" by extensive unresolved transitions. Reliably modeling such multi-scale structures is a significant computational challenge.



At high densities, shielding by and collisions with free electrons lead to density broadening, pressure ionization, and an explosion of statistically accessible multiply excited states.

11 Bailey *et al.* PRL **99**, 265002 (2007).

Recent experiments on LCLS brought about a controversy on Ionization Potential Depression (IPD)

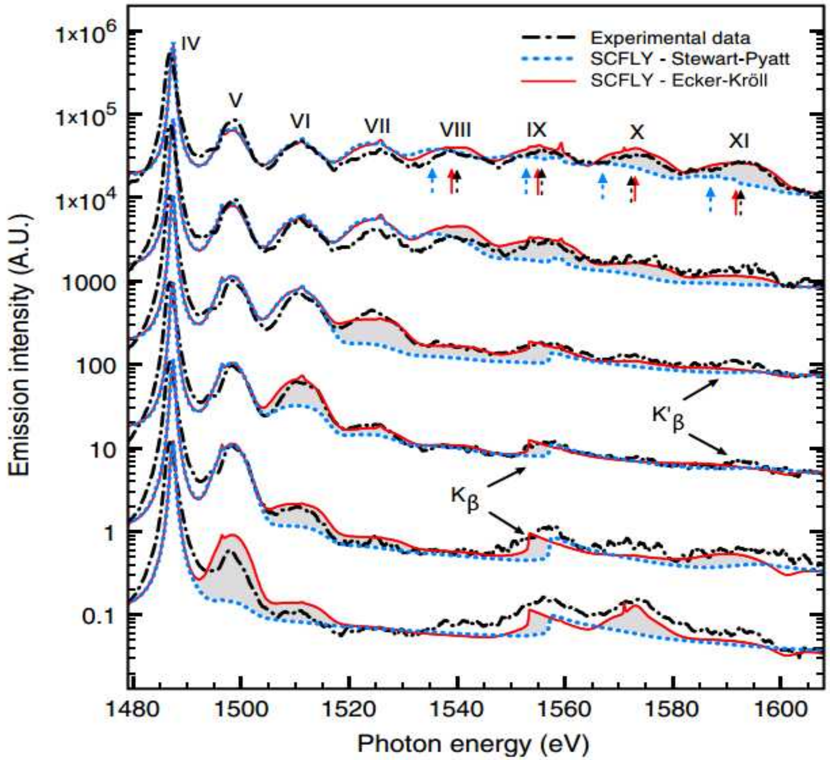


Fig. 1 from Ciricosta, Vinko, Chung *et al.*, PRL **109** 065002 (2012).

At least four different explanations have been advanced, each of which claims to account for the entire discrepancy between EK and SP:

1. B. Crowley, "Continuum Lowering – A New Perspective," *HEDP* (2014).
2. Son, Thiele, Jurek *et al*., "Quantum-Mechanical Calculation of Ionization-Potential Lowering in Dense Plasmas," PRX (2014).
3. Vinko, Ciricosta, & Wark, "Density functional theory calculations of continuum lowering in strongly coupled plasmas," *Nature Comm.* (2014).
4. Iglesias, "A plea for a reexamination of ionization potential depression measurements," *HEDP* (2014).

As yet, no model with the fine detail required to produce a credible spectrum for comparison with data has a consistent treatment of density and kinetic effects [4].

Summary: Consistency is a key component of reliable predictions in strongly coupled systems

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- Bulk thermal and electrical transport:
  - Critical for hydrodynamic simulations
  - Widely used Ziman formulation depends on DOS, $d\sigma/d\Omega$, and S(k)
  - Errors can arise if these are not all computed in the same basis

- X-ray Thomson Scattering:
  - Has become a popular diagnostic for warm dense matter
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Opinion: model development should emphasize internal consistency
and seek comparisons with experimental data wherever possible – QMD can help here.

Ionic component: the structure factor $S(q)$ has a large effect on η in strongly coupled systems

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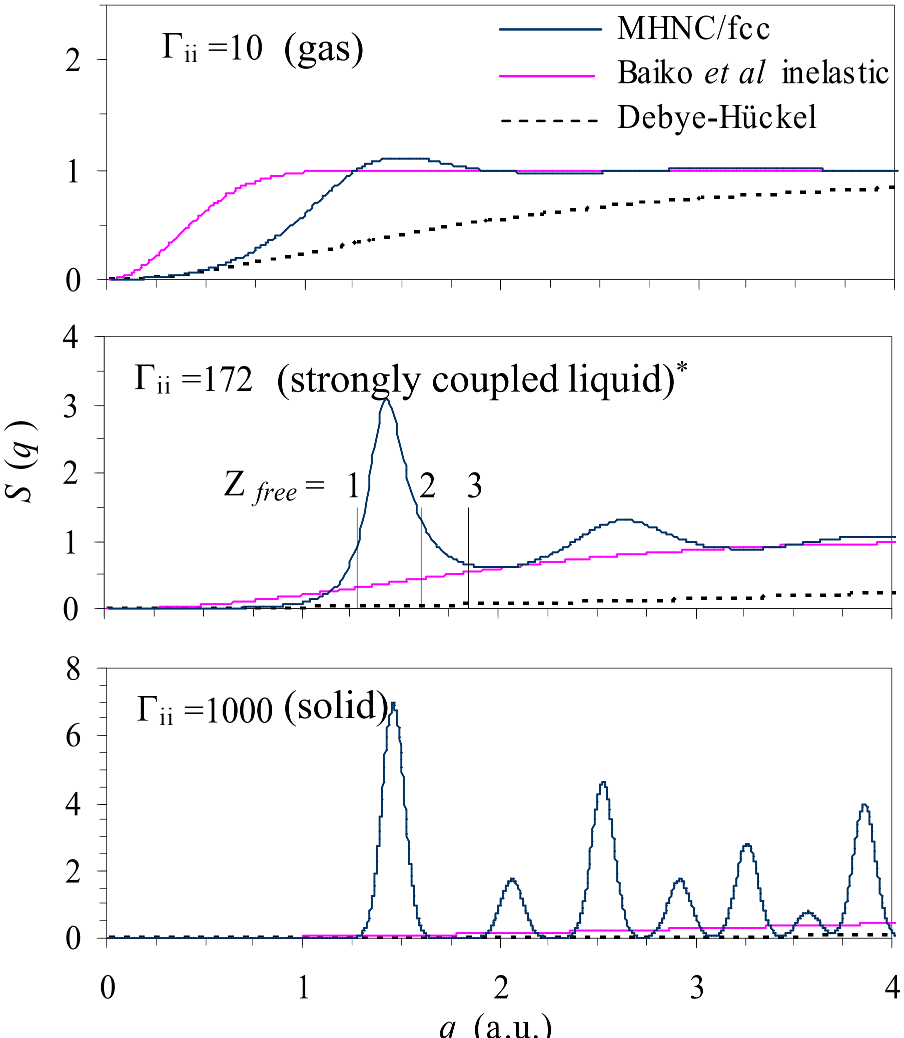

*Young, Corey, and DeWitt, PRA **44**, 6508 (1991).

$$\eta = -\frac{\Omega}{3\pi}\frac{1}{Z_0 Z_i}\int_0^\infty \frac{\partial f(\varepsilon)}{\partial \varepsilon}\left\{\int_0^{2p} q^3\left(\frac{d\sigma(p,\theta)}{d\theta}\right)S(q)\,dq\right\}d\varepsilon$$

$S(q)$ is the Fourier transform of the ion-ion correlation function $g(r)$, which gives the probability of encountering an ion at a given radius.

$S(q)$ varies with the ion-ion coupling parameter $\Gamma_{ii} = Z_0^2/TR_0$:

For solids, $S(q)$ has well-defined Bragg peaks. As the temperature increases, the peaks broaden due to lattice vibrations (phonons).

In the ideal gas limit, where ions are weakly coupled, $S(q) \rightarrow 1$.