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Ultrafast X-Ray Absorption Spectroscopy of Warm Dense Matter

Steve Johnson (PSI, Switzerland)

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Co-workers

- A. M. Lindenberg,* A. G. MacPhee, R. W. Falcone (UC Berkeley)
- P. A. Heimann, O. Monteiro (LBNL)
- R. W. Lee (LLNL)
- J. J. Rehr (Univ. of Washington)
- Z. Chang (Kansas State University)
- H. O. Jeschke (Rutgers)
- M. E. Garcia (Freie Universität Berlin)

* now at SSRL

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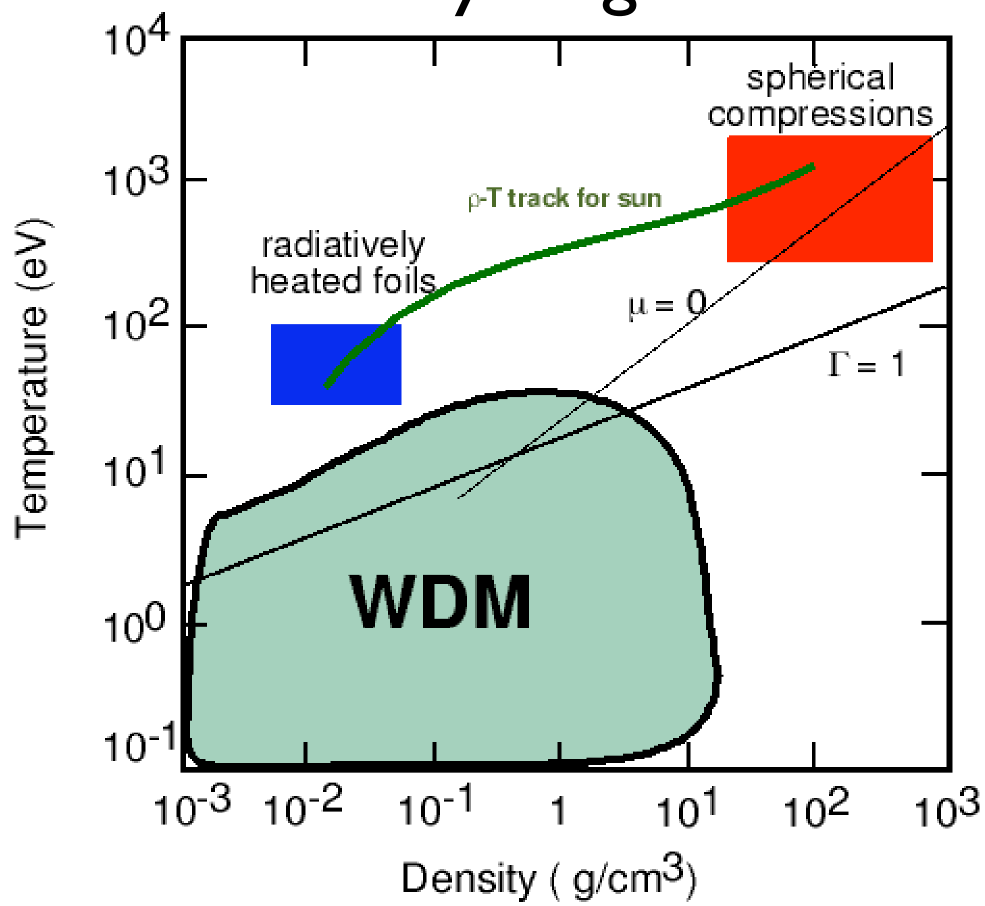
Outline

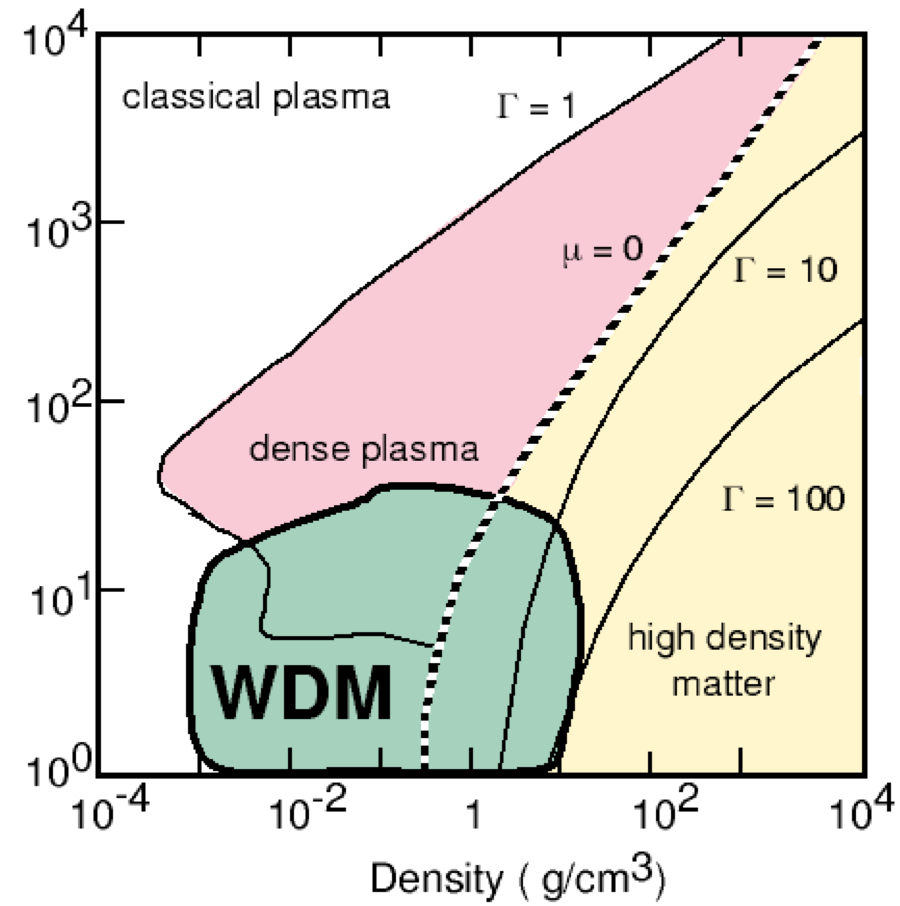
- Overview: warm dense matter
- Technique: picosecond XAS
- Results:
  - Liquid silicon
  - Liquid carbon
- Future: hard X-rays at SLS

Warm dense matter

Hydrogen



Aluminum



- T ~ 1-10 eV (10^4-10^5 K)   ρ ~ 10^{-3}-10 g/cm^3
- Conditions found in explosions, astrophysics

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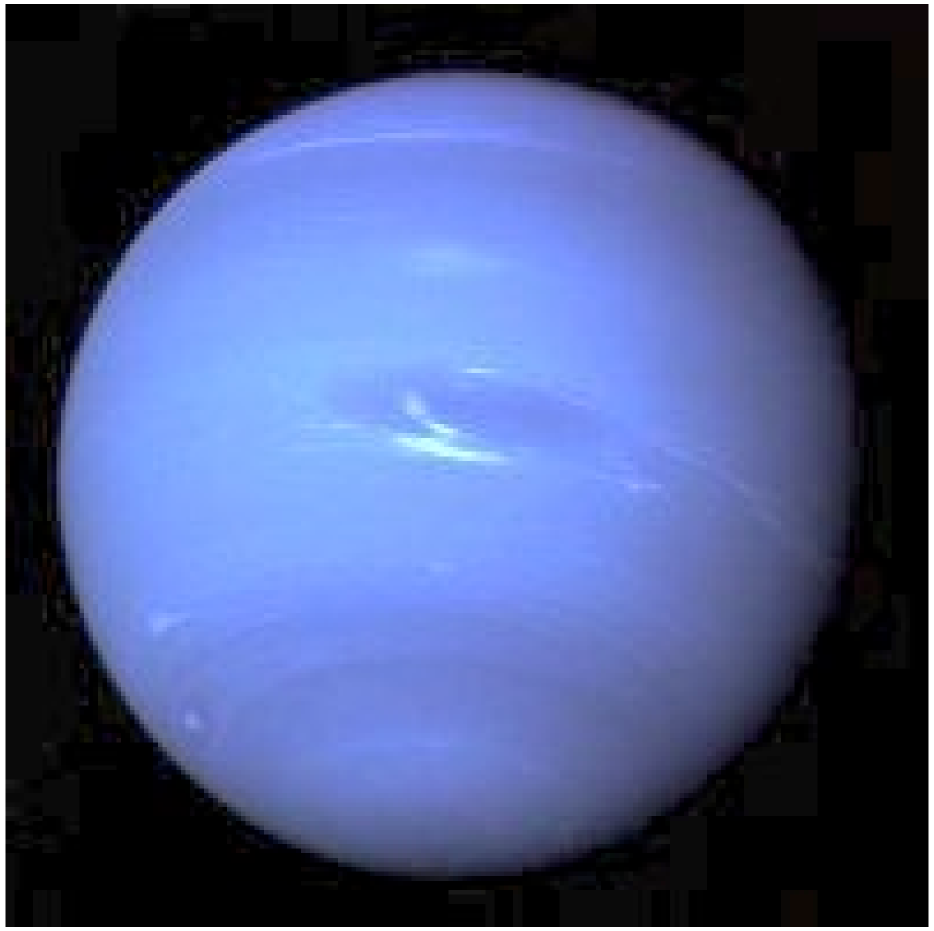
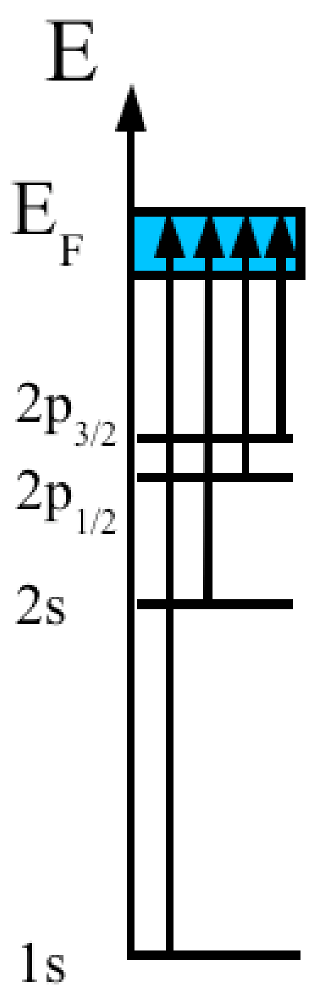
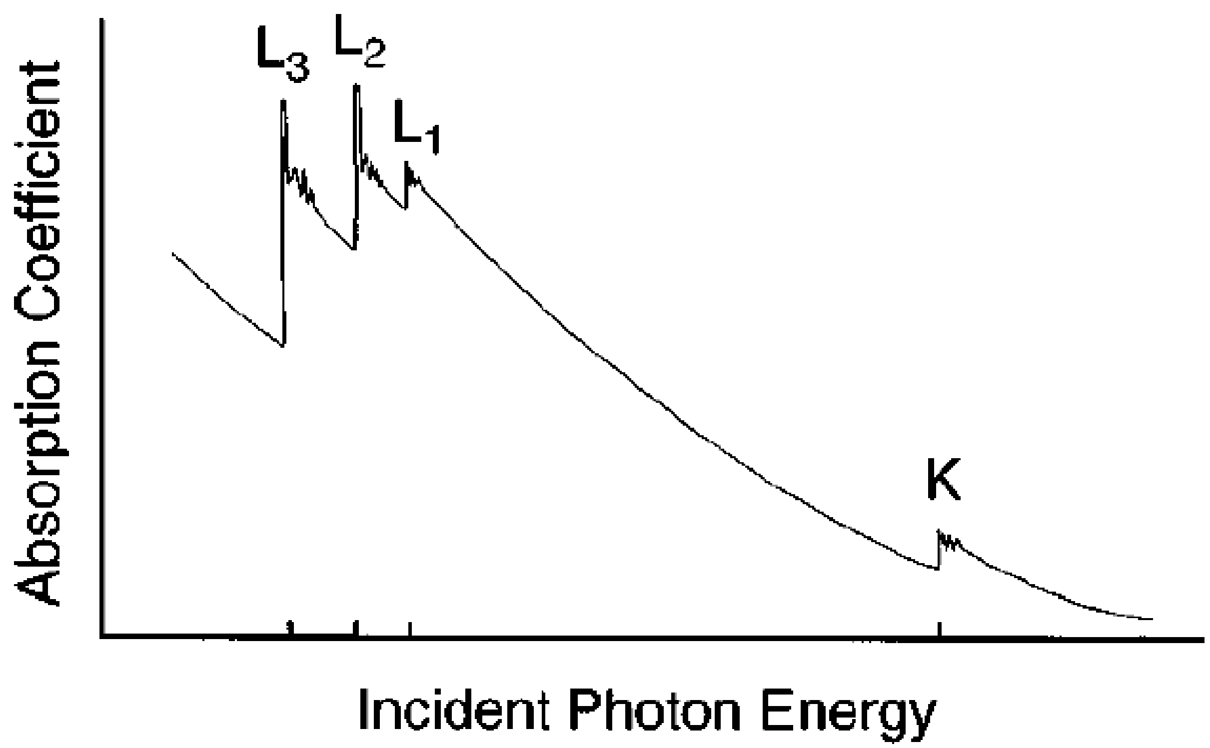
Warm dense matter

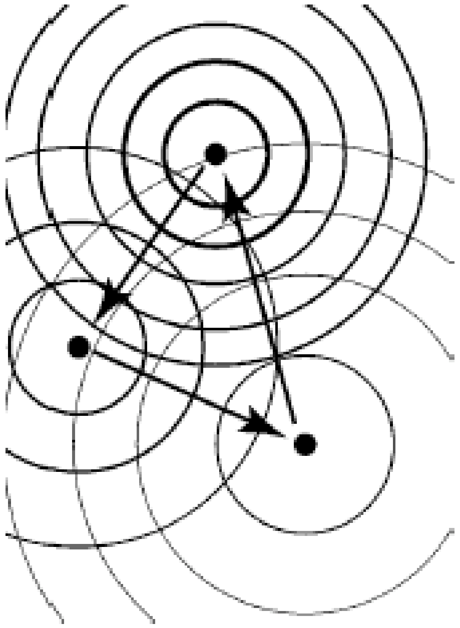


- WDM difficult to maintain here on earth
- Volatility requires time-resolved technique

Technique: ps-XAS

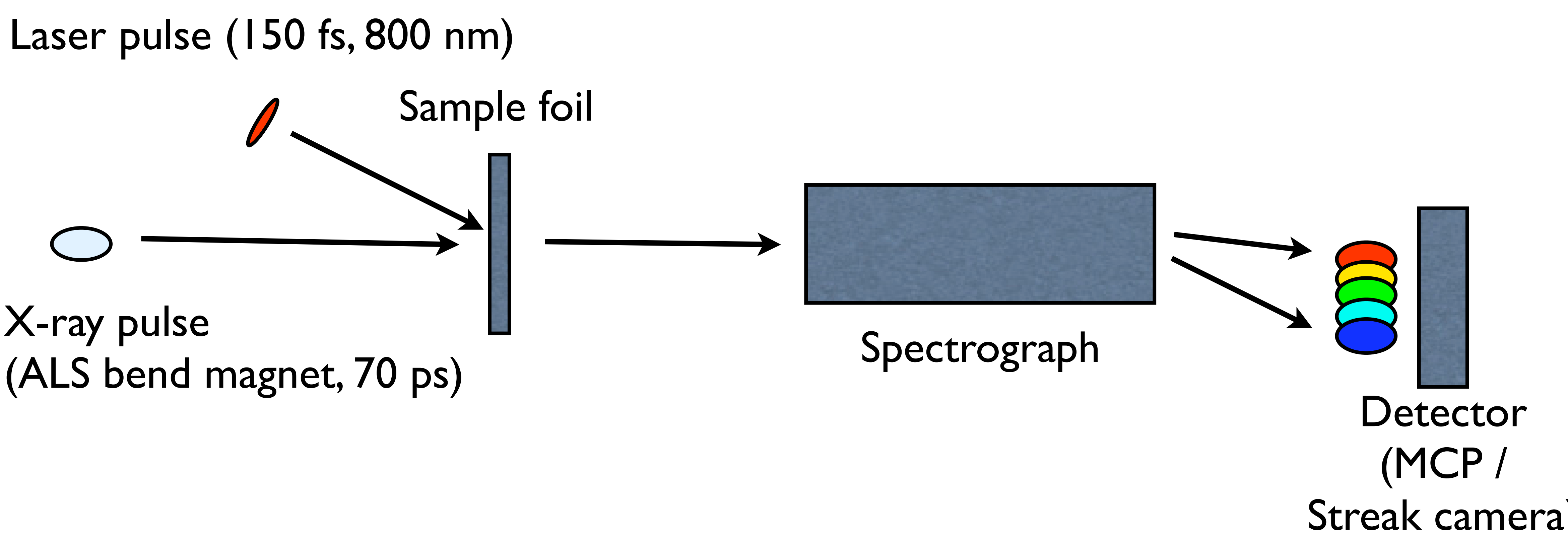






- Near-edge → electronic structure
- EXAFS → atomic structure

Technique: ps-XAS



- Rapidly heat sample with fs laser pulse
- X-rays probe before sample evaporates/expands

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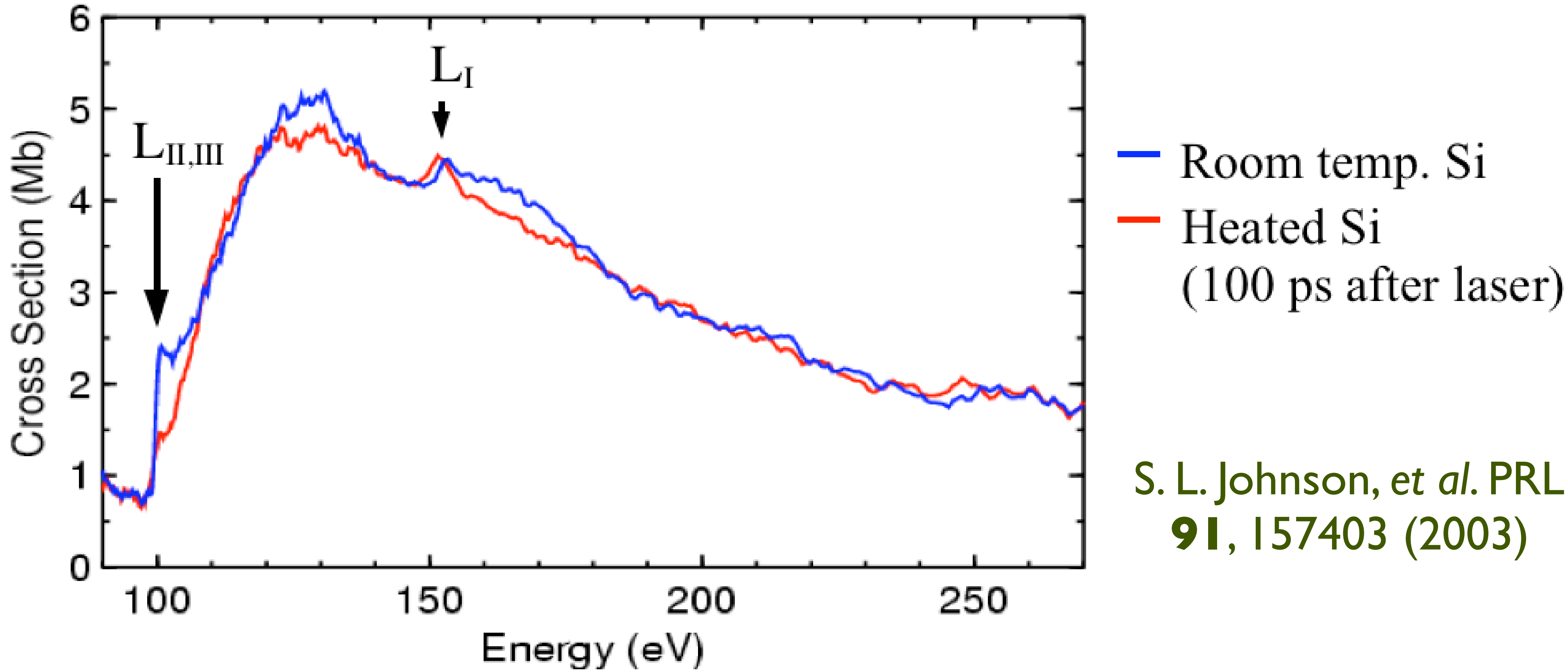
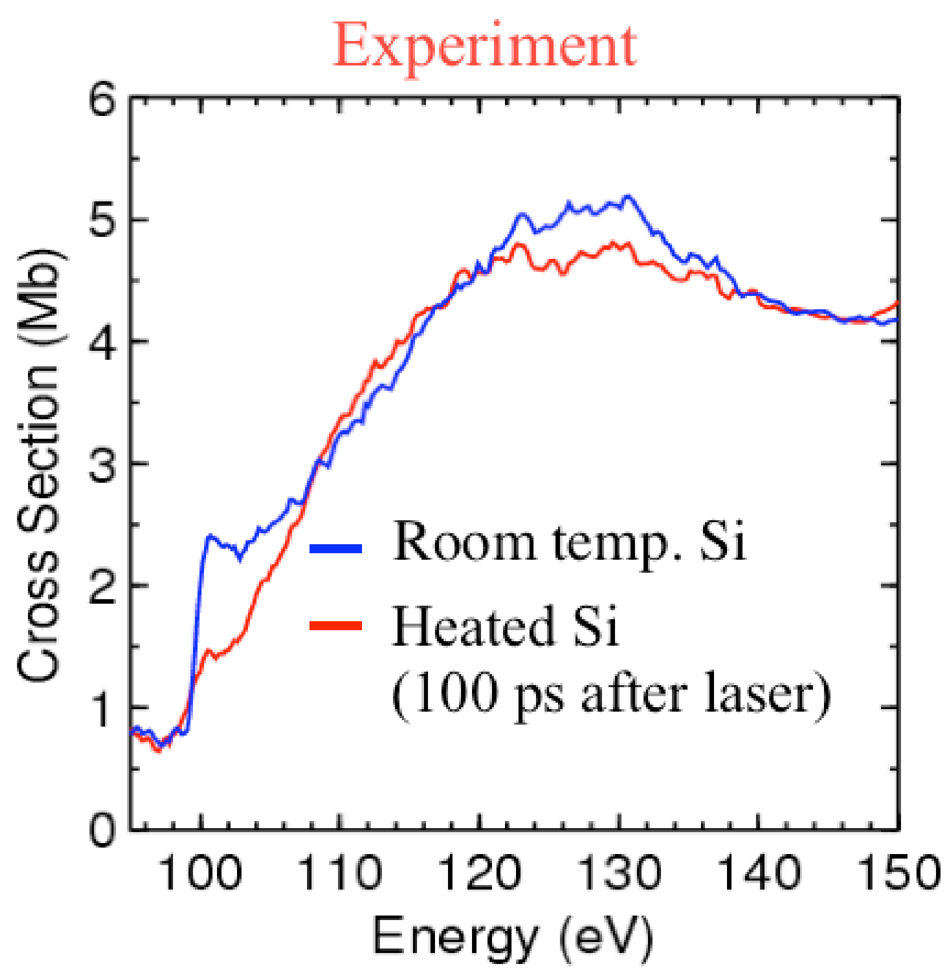
Liquid silicon

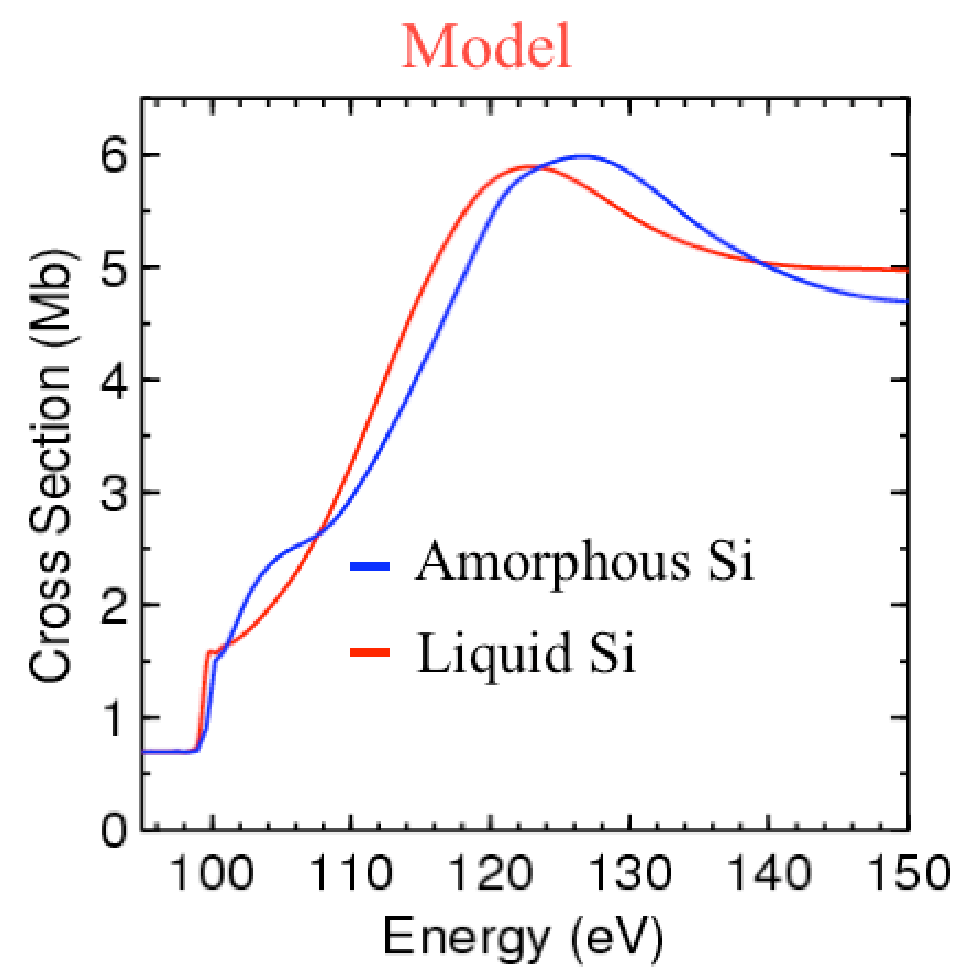


S. L. Johnson, *et al.* PRL **91**, 157403 (2003)

- First experiment: liquid Si near T_c (~5000 K)
- Large changes from solid

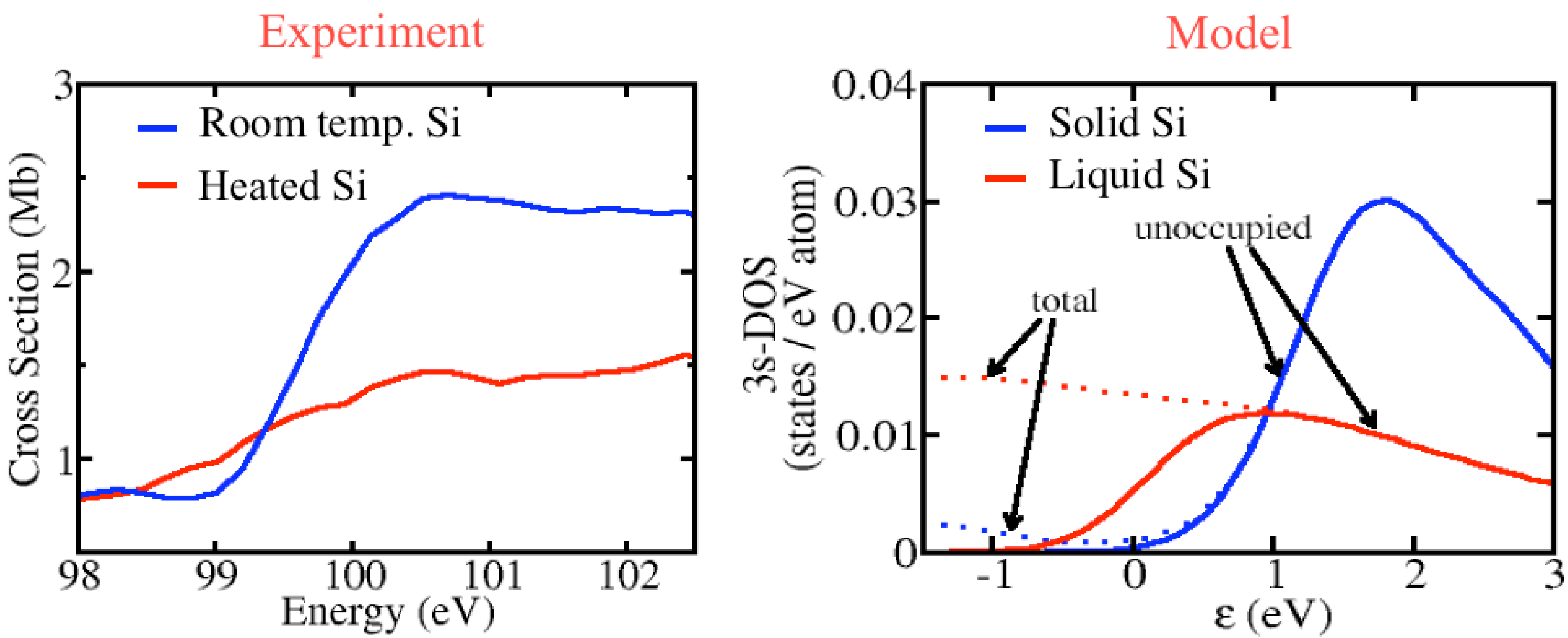
Liquid silicon





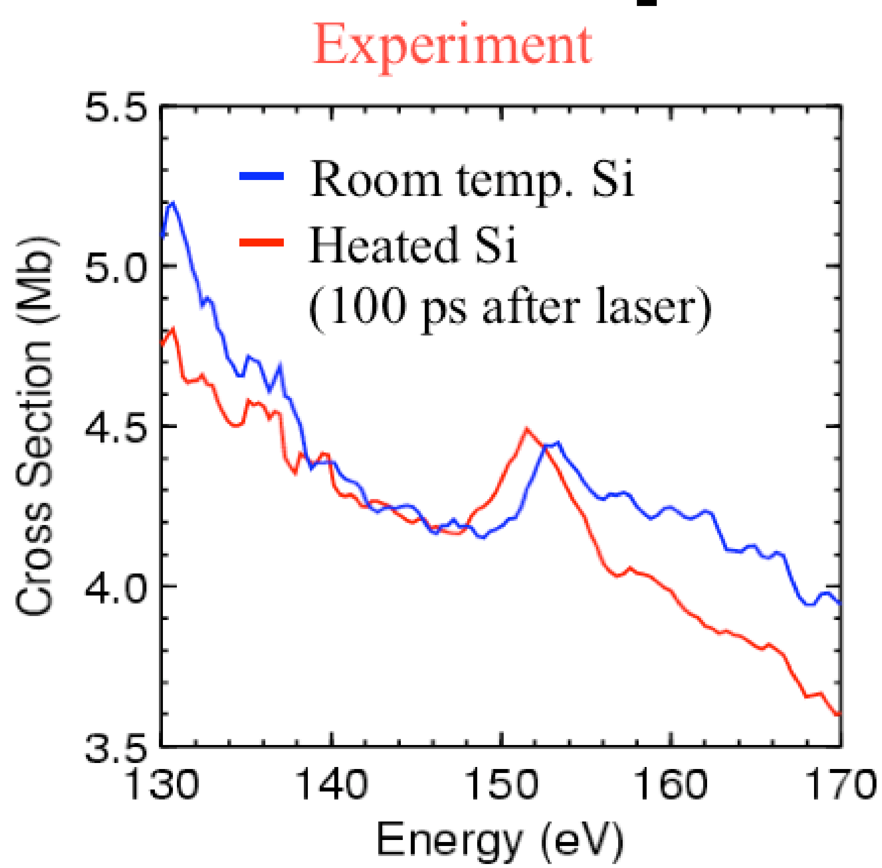
- MD simulations + FEFF x-ray code → model
- Shift in p-to-d peak → (0.15 ± 0.07) Å increase in nearest-neighbor distance

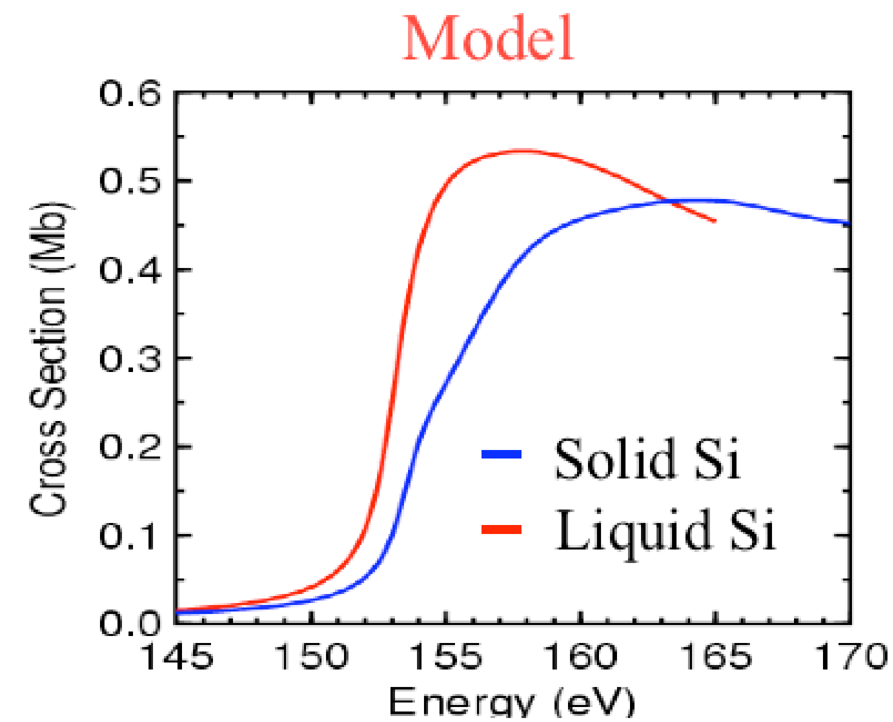
Liquid silicon

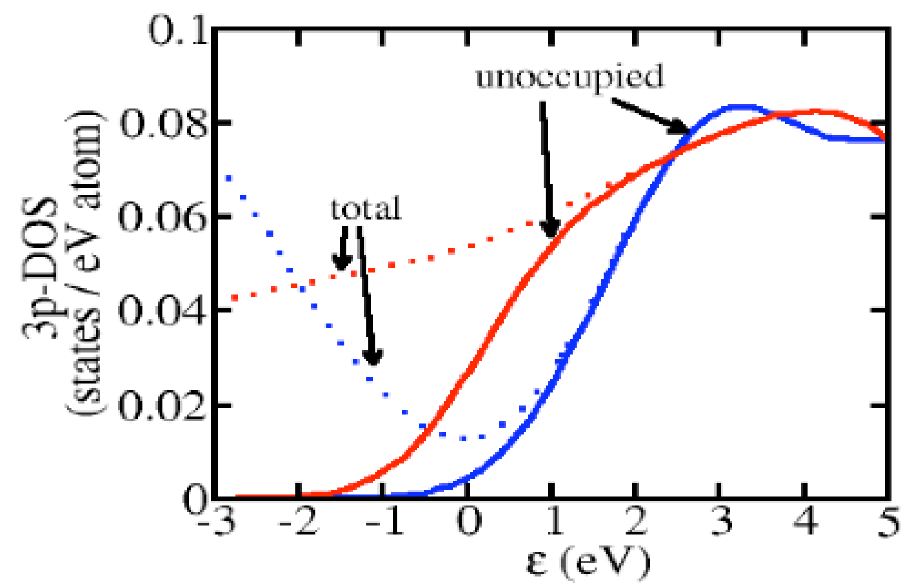


- Region closer to $L_{II,III}$ edge better modeled by 3s-DOS
- Decrease in edge jump related to band gap collapse

Liquid silicon

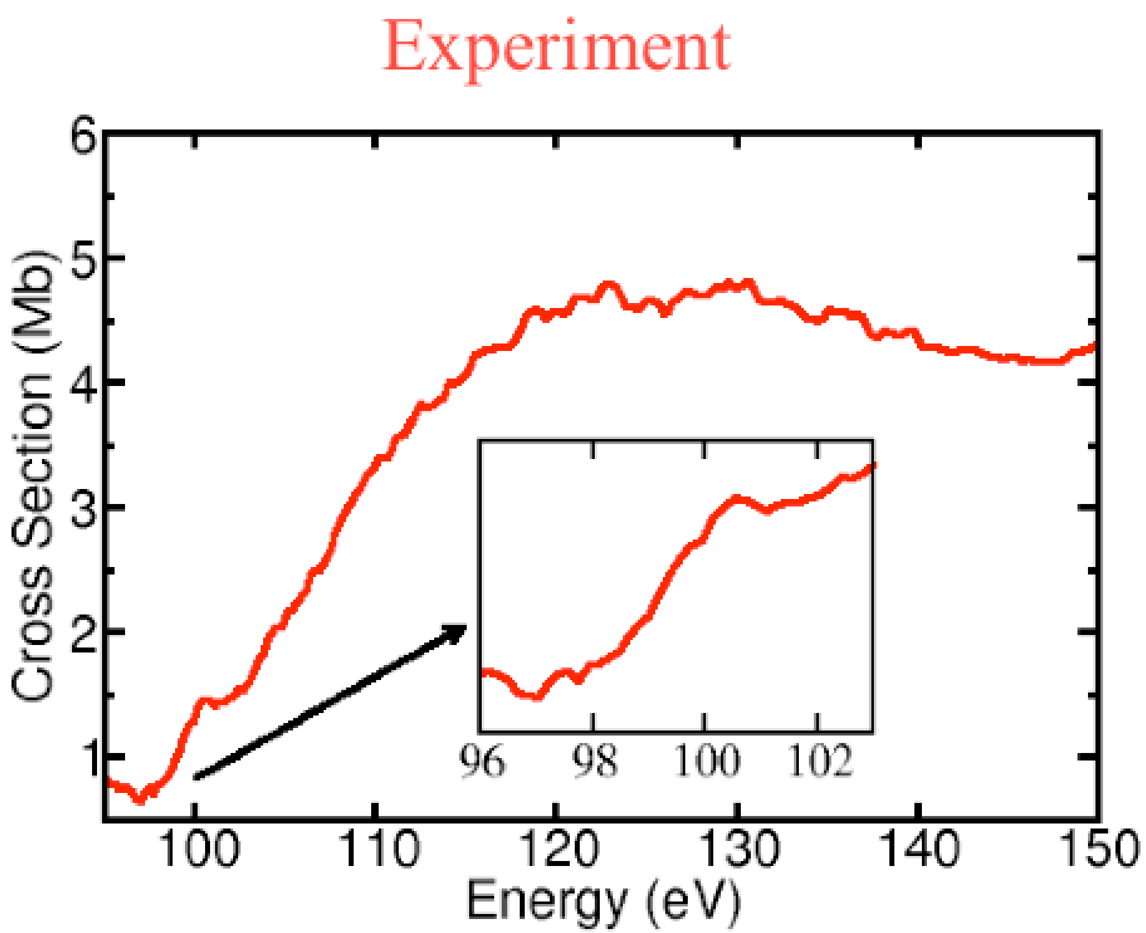


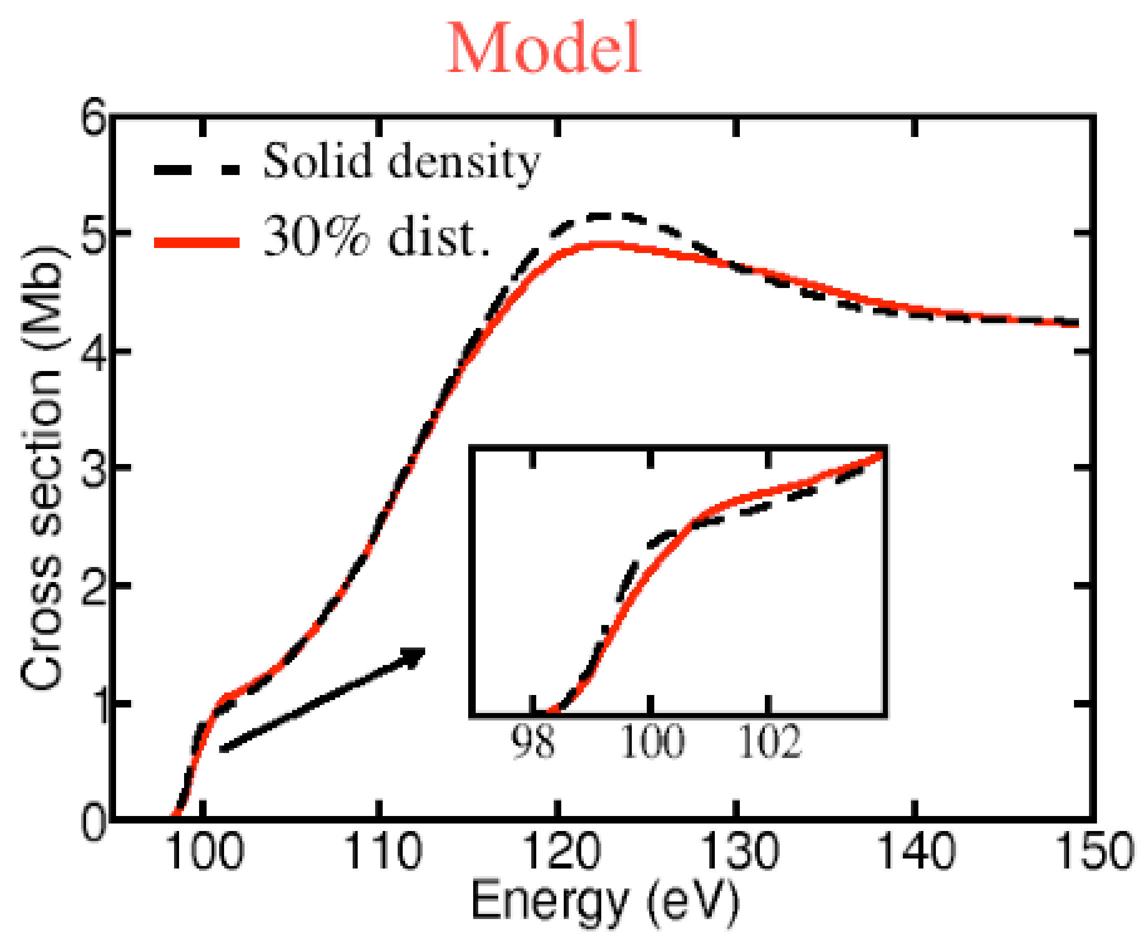




- L_I edge: experiment sees (-1.6 ± 0.2) eV shift
- Model predicts -1.2 eV shift from band gap collapse, DOS changes

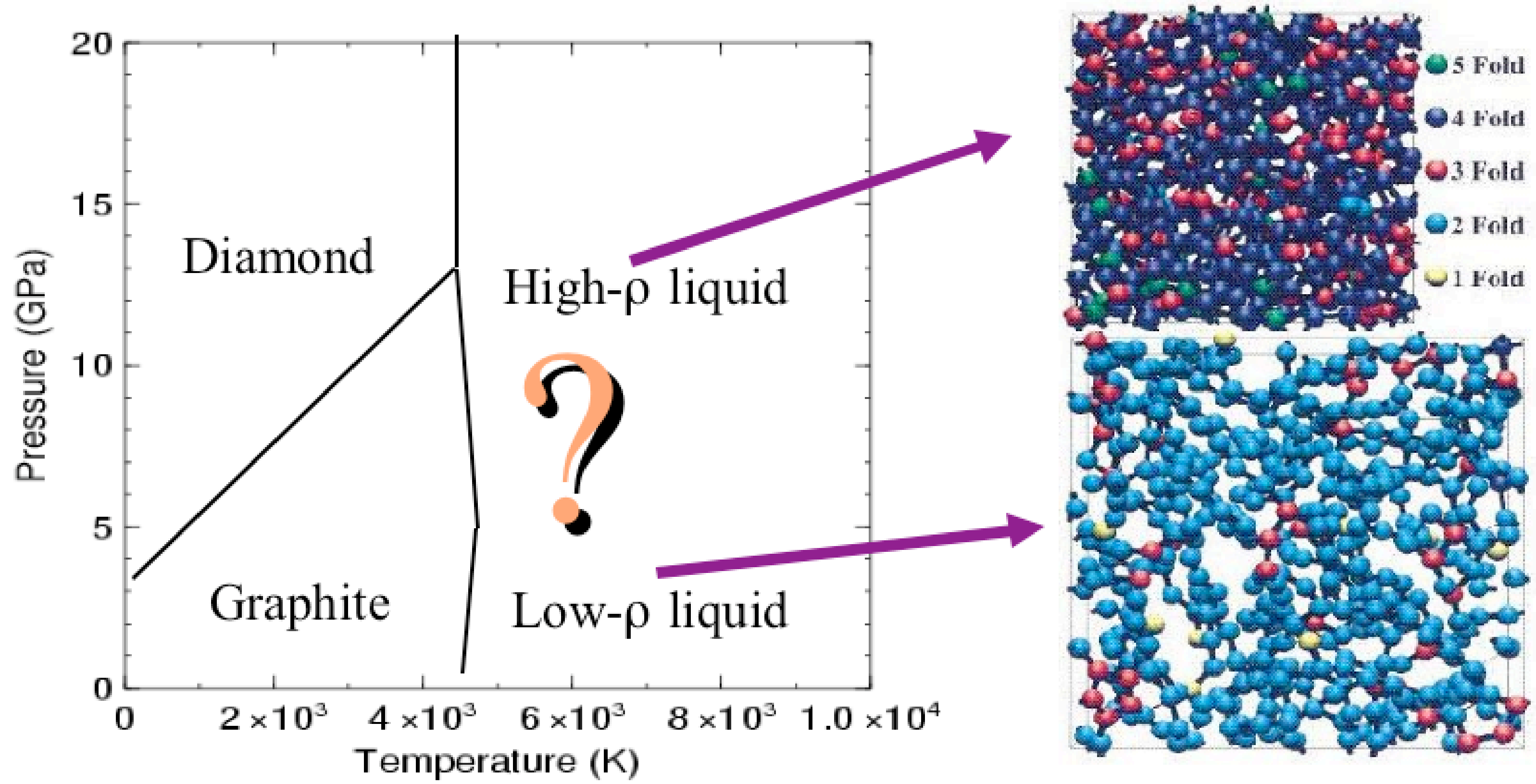
Liquid silicon





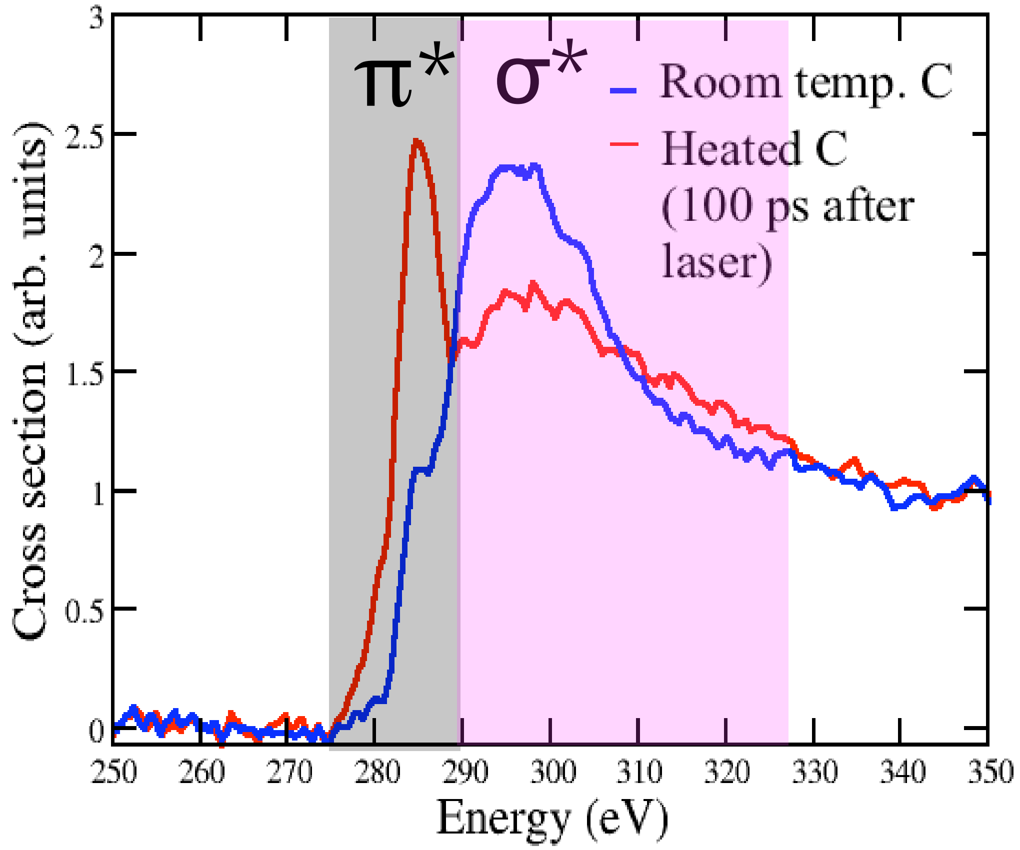
- In general, experimental data show more broadening than expected
- May be due to local density variations near T_c

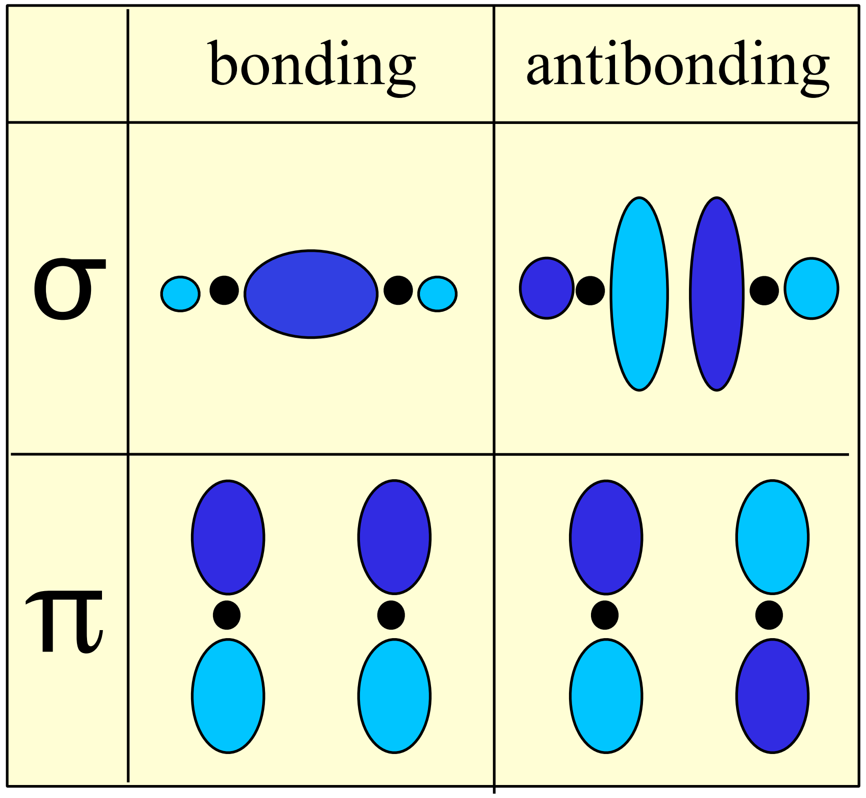
Liquid carbon



- Difficult system due to high T_m
- Important for astrophysics: Uranus, Neptune
- Intermediate in nanotube synthesis: W. de Heer, *Science* 307, p. 309 (2005)

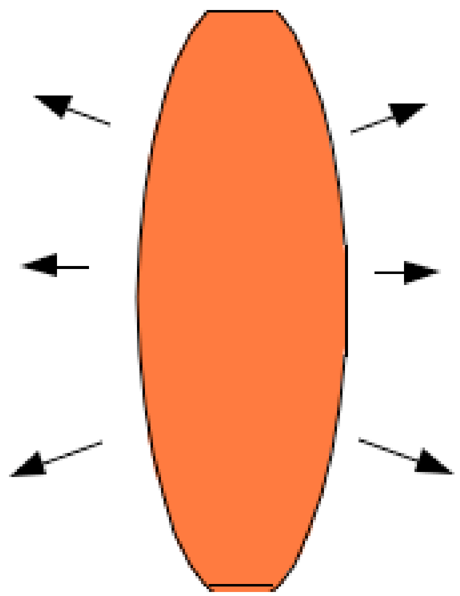
Liquid carbon

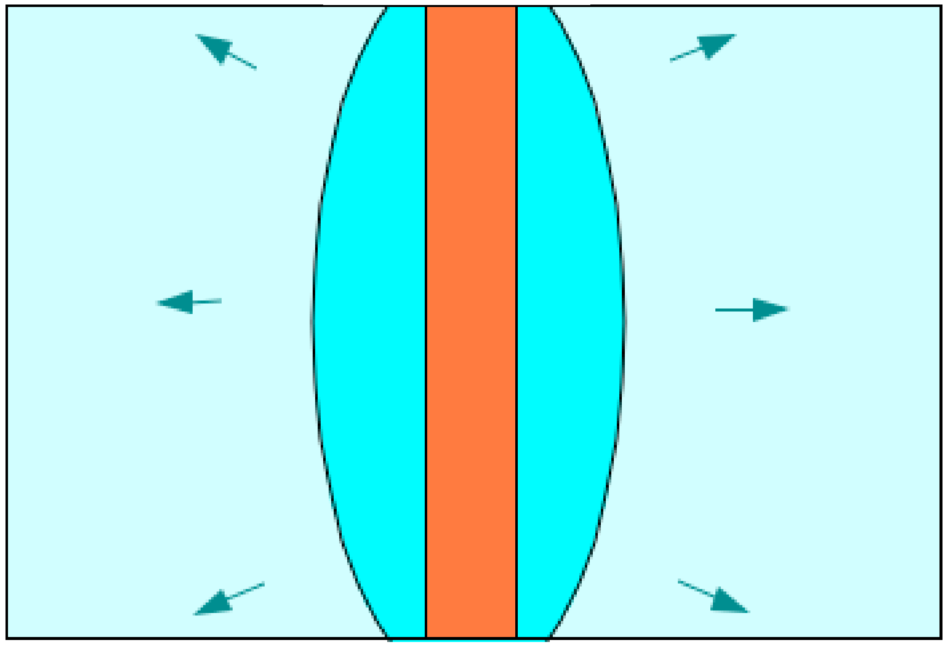




- Melting of free-standing a-C foil → big changes in antibonding states
- Increase in π* states, decrease in σ* states

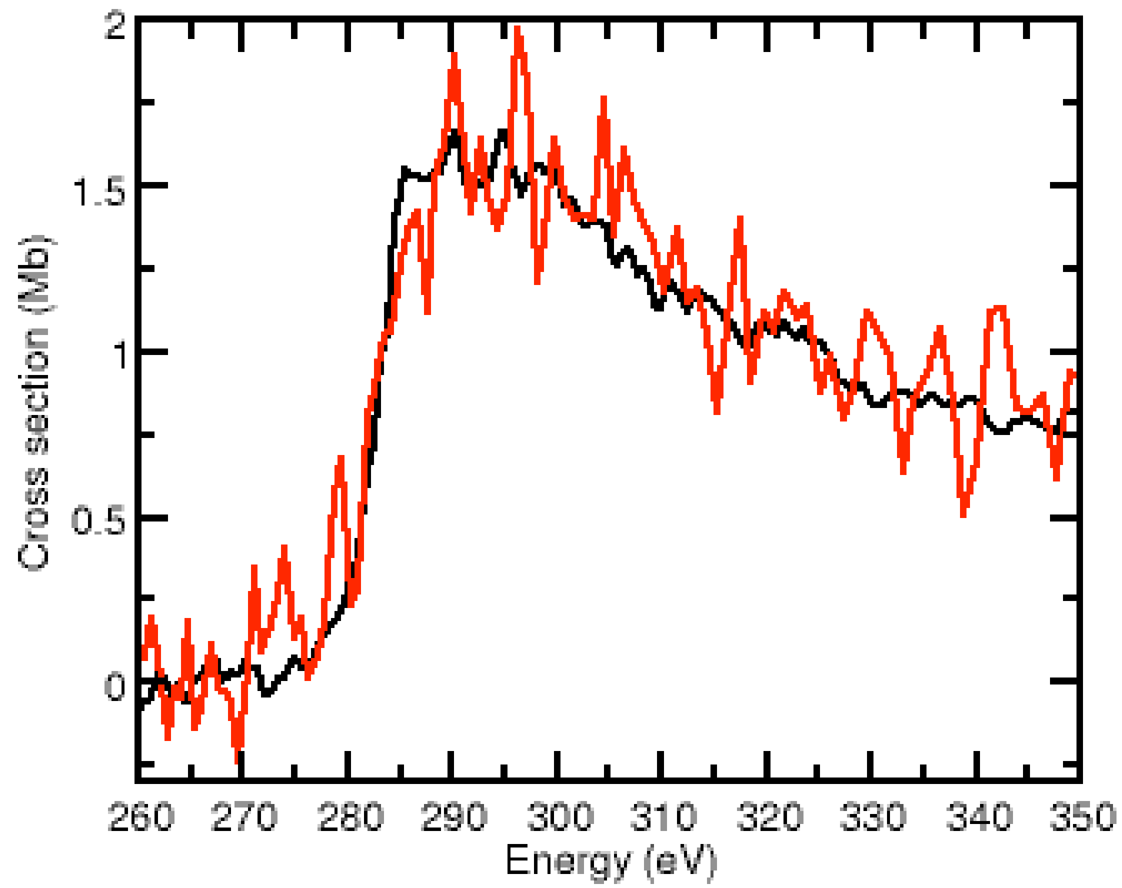
Liquid carbon





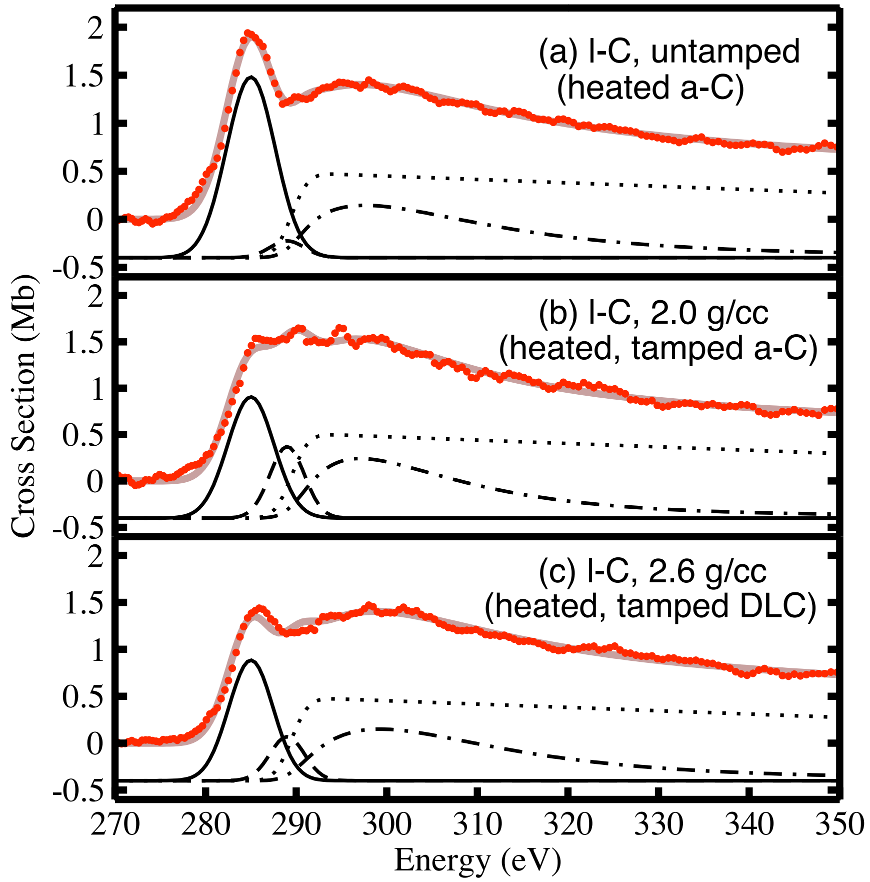
- Higher densities: tamping with LiF
- Expansion delayed by ~100 ps

Liquid carbon



- Comparison: tamped a-C at 100 ps (black) with free-standing a-C at 5 ps (red, measured with streak camera)
- Tamping does appear to delay expansion

Liquid carbon

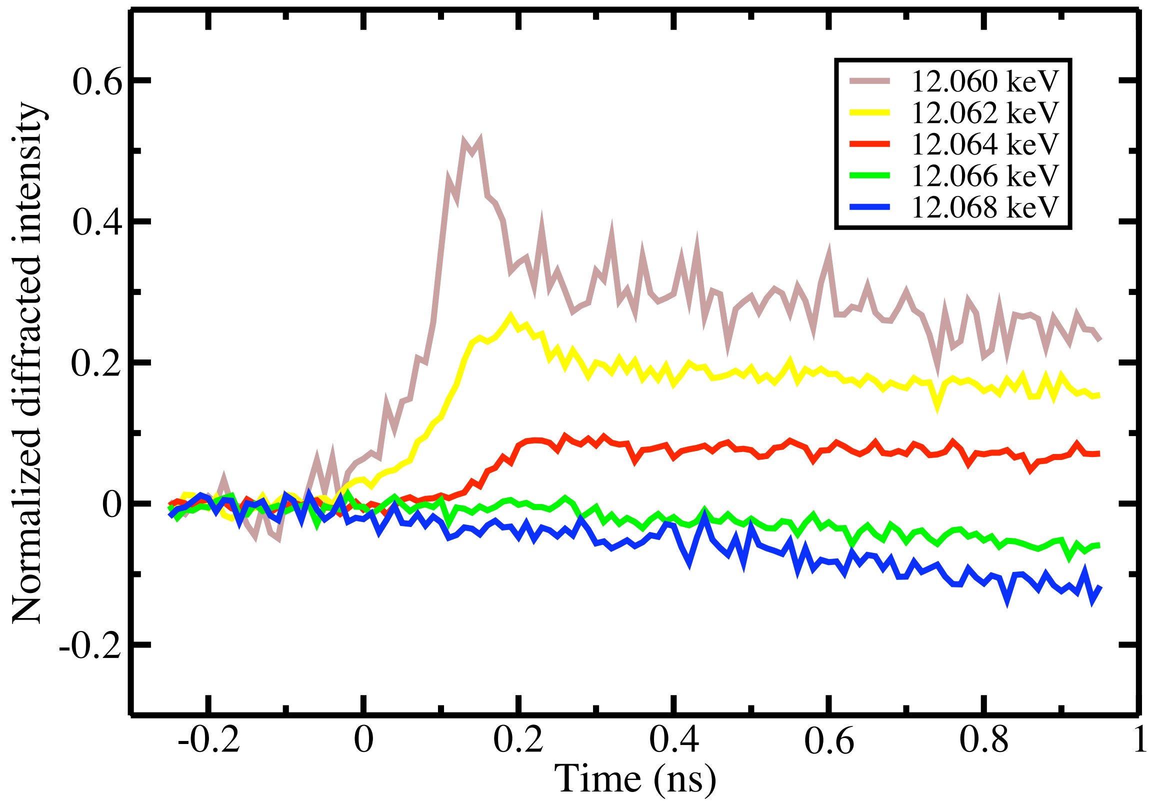


Material	π^* states/site (fit)	π^*/atom (sim.)	σ_c^* peak (eV)
a-C	$0.7^{+0.2}_{-0.1}$	···	295.5 ± 0.2
DLC	$0.4^{+0.2}_{-0.1}$	···	295.6 ± 0.3
l-C, untamped	$2.3^{+0.1}_{-0.5}$	···	297.6 ± 0.9
l-C, 2.0 g/cm^3	$1.5^{+0.2}_{-0.3}$	1.5 ± 0.1	297.1 ± 0.7
l-C, 2.6 g/cm^3	$1.4^{+0.2}_{-0.3}$	1.2 ± 0.1	299.2 ± 0.9

S. L. Johnson, *et al.* PRL **94**, 057407 (2005)

- Curve-fitting gives estimate of π* states/site
- Low density is sp-bonded, higher densities a mixture
- Agrees with tight-binding MD models

Future: hard X-rays



- Develop ps- and fs-XAS capability for photon energies 5-15 keV
- Invesitigate transition to WDM (melting)
- Other high energy-density systems

Conclusions

- Time-resolved XAS: good tool for WDM
- Liquid silicon XAS shows good agreement with MD models, except for broadening
- Liquid carbon is sp-bonded at low densities, a mixture at higher densities
- Work on extending the technique to hard X-rays in progress at SLS