Ultrafast X-Ray Absorption Spectroscopy of Warm Dense Matter

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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

- $T \sim 1-10$ eV ($10^4-10^5$ K) \quad $\rho \sim 10^{-3}-10$ g/cm$^3$
- Conditions found in explosions, astrophysics
Warm dense matter

- WDM difficult to maintain here on earth
- Volatility requires time-resolved technique
Technique: ps-XAS

- Near-edge $\rightarrow$ electronic structure
- EXAFS $\rightarrow$ atomic structure
Technique: ps-XAS

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

- First experiment: liquid Si near $T_c$ ($\sim 5000$ 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

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

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In general, experimental data show more broadening than expected.

May be due to local density variations near $T_c$. 

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Liquid carbon

- Difficult system due to high $T_m$
- Important for astrophysics: Uranus, Neptune
Liquid carbon

- Melting of free-standing a-C foil $\rightarrow$ big changes in antibonding states
- Increase in $\pi^*$ states, decrease in $\sigma^*$ 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

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

<table>
<thead>
<tr>
<th>Material</th>
<th>$\pi^*$ states/site (fit)</th>
<th>$\pi^*/$atom (sim.)</th>
<th>$\sigma^*$ peak (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$-C</td>
<td>$0.7^{+0.2}_{-0.1}$</td>
<td>$\cdots$</td>
<td>$295.5 \pm 0.2$</td>
</tr>
<tr>
<td>DLC</td>
<td>$0.4^{+0.2}_{-0.1}$</td>
<td>$\cdots$</td>
<td>$295.6 \pm 0.3$</td>
</tr>
<tr>
<td>$l$-C, untamped</td>
<td>$2.3^{+0.1}_{-0.5}$</td>
<td>$\cdots$</td>
<td>$297.6 \pm 0.9$</td>
</tr>
<tr>
<td>$l$-C, 2.0 g/cm$^3$</td>
<td>$1.5^{+0.2}_{-0.3}$</td>
<td>$1.5 \pm 0.1$</td>
<td>$297.1 \pm 0.7$</td>
</tr>
<tr>
<td>$l$-C, 2.6 g/cm$^3$</td>
<td>$1.4^{+0.2}_{-0.3}$</td>
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<td>$299.2 \pm 0.9$</td>
</tr>
</tbody>
</table>

Future: hard X-rays

- Develop ps- and fs-XAS capability for photon energies 5-15 keV
- Investigate 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