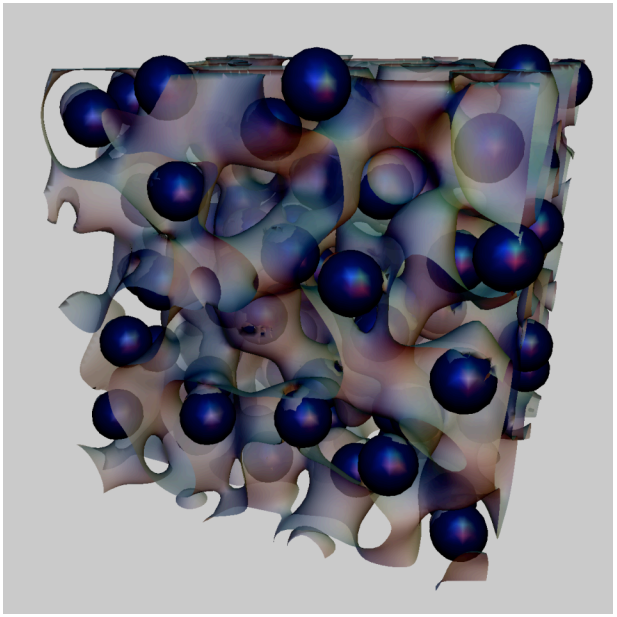
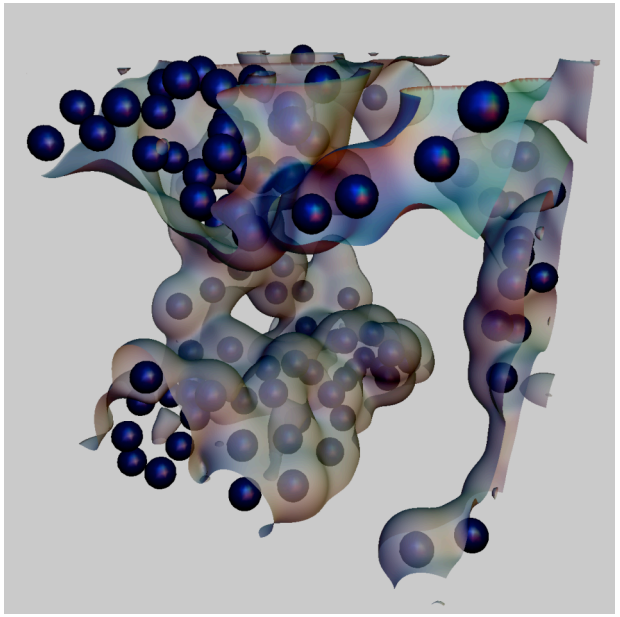
SAND2006-7376P

Quantum Molecular Dynamics Calculation of Electrical and Thermodynamic Properties for High Energy Density Physics





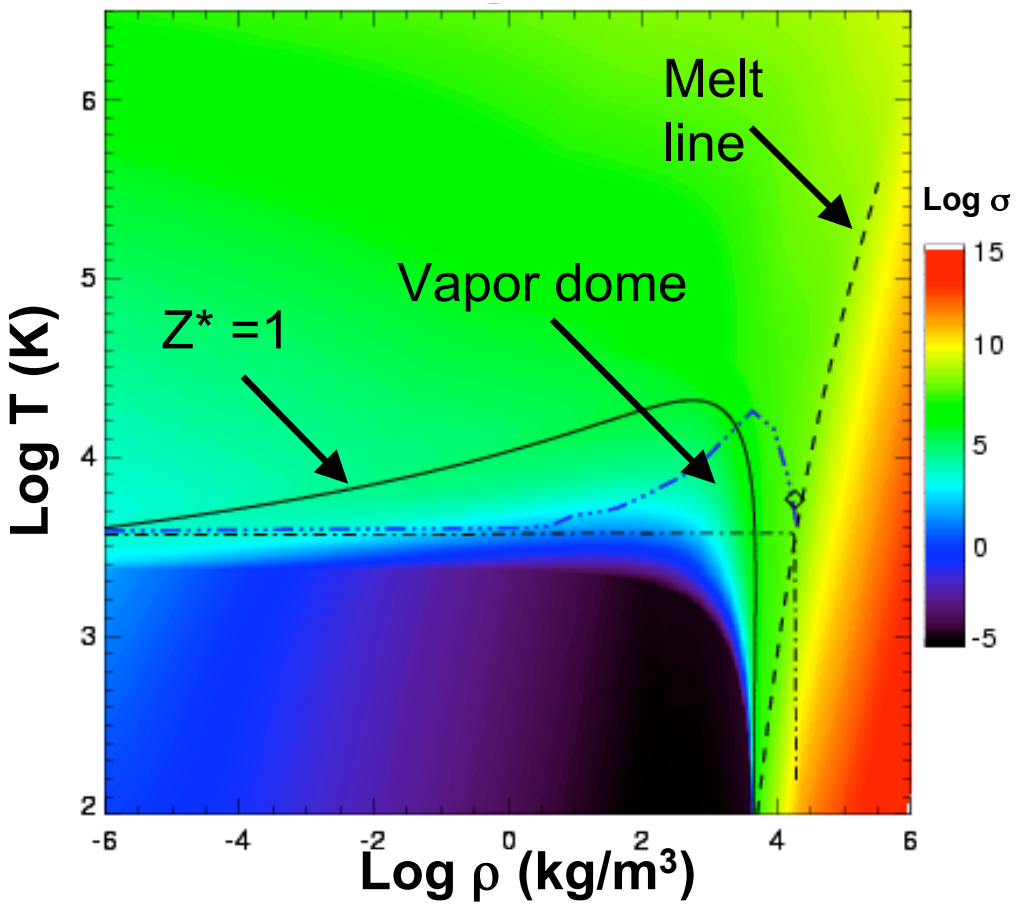
Mike Desjarlais
HEDP Theory &
ICF Target Design
Sandia National Laboratories

Briefing to VNIIEF Visitors
Albuquerque, New Mexico
November 14, 2006

Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

HEDP computer simulations rely on "physics packages": Conductivities, Equations of State, and Opacities

Tungsten Conductivity



This highly structured portion of phase space is Warm Dense Matter

Definitions of Warm Dense Matter are varied, but generally center around **strongly coupled ions and moderately degenerate electrons** --- many different interactions are comparable.

A **quantum mechanical** treatment is generally necessary.

Most wide-range equations of state interpolate through this difficult area.

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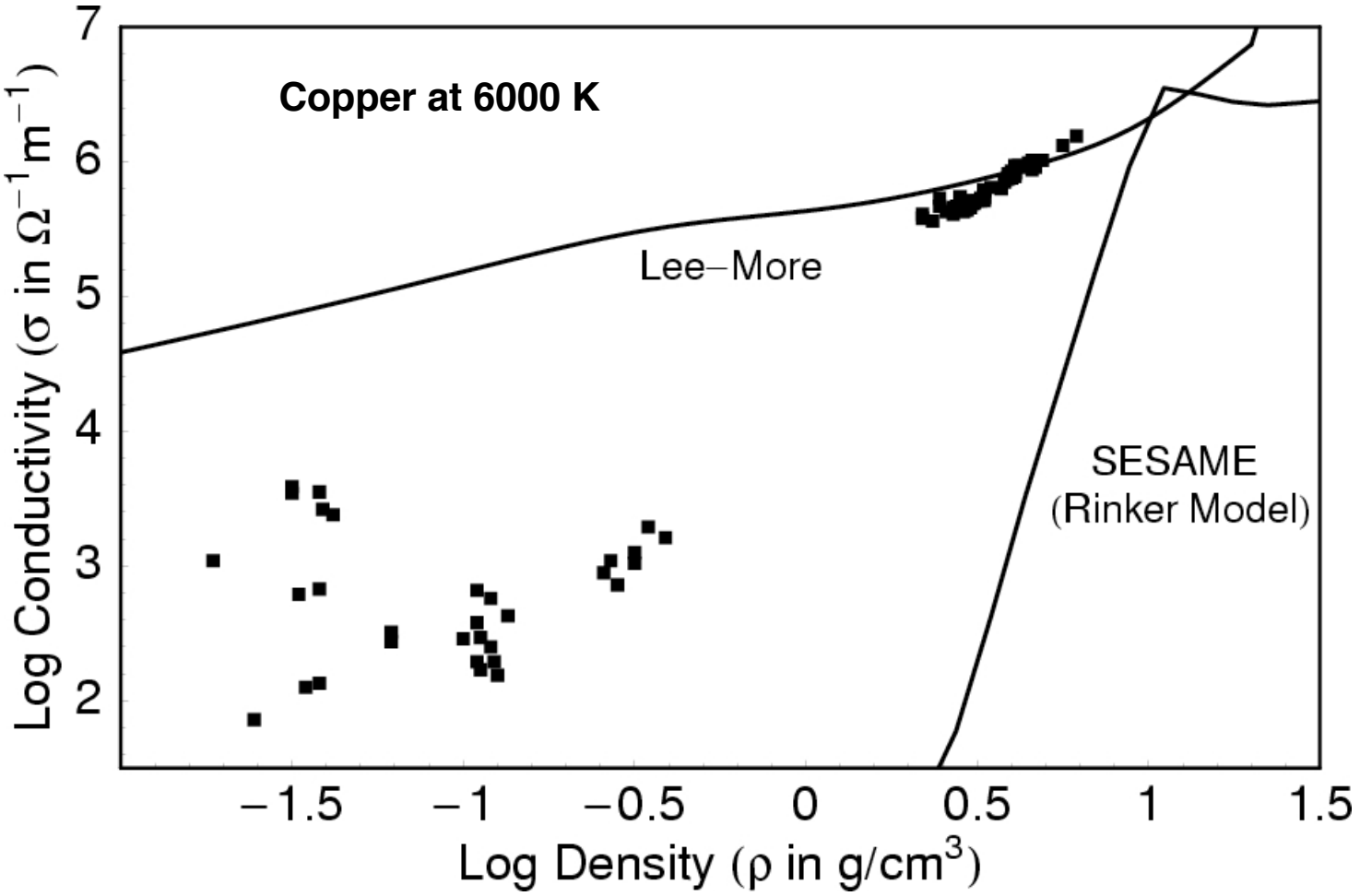
The Lee-More and SESAME (Rinker) models are inaccurate near the metal-insulator transition



Data courtesy of Alan DeSilva, University of Maryland

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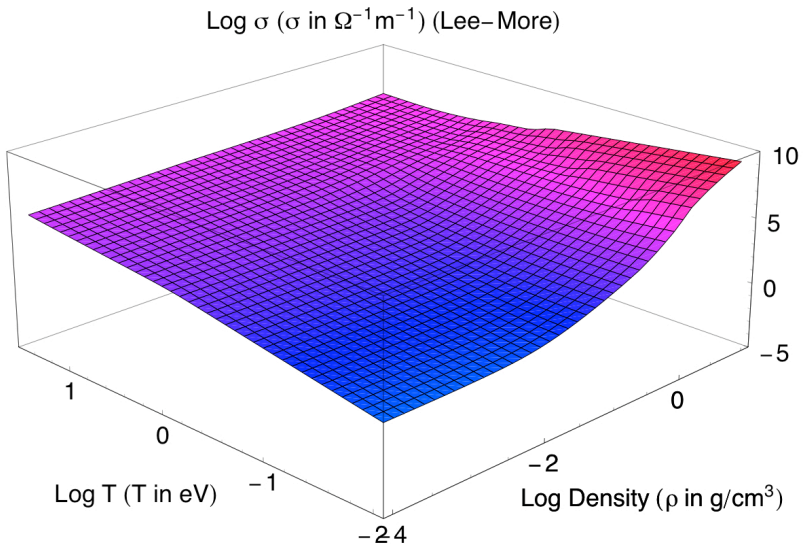
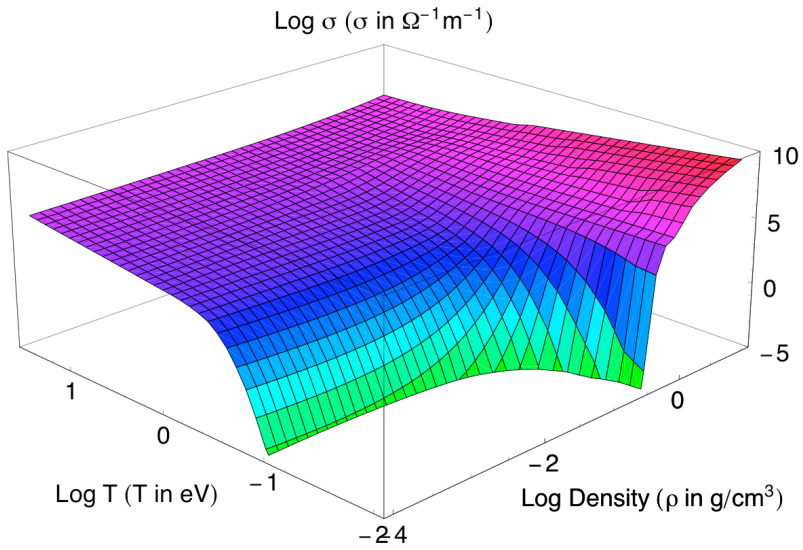
Modifications of the Lee-More algorithm were made to obtain an improved wide-range model*

* M. P. Desjarlais, Contrib. Plasma Phys. **41** (2001) 2-3, 267-270





Principal modifications:

$$\sigma = n_e A(\frac{\mu}{kT}, \omega\tau)\frac{e^2}{m}\tau$$

Ionization equilibrium model, pressure ionization

Electron-neutral collisions, Minimum collision time

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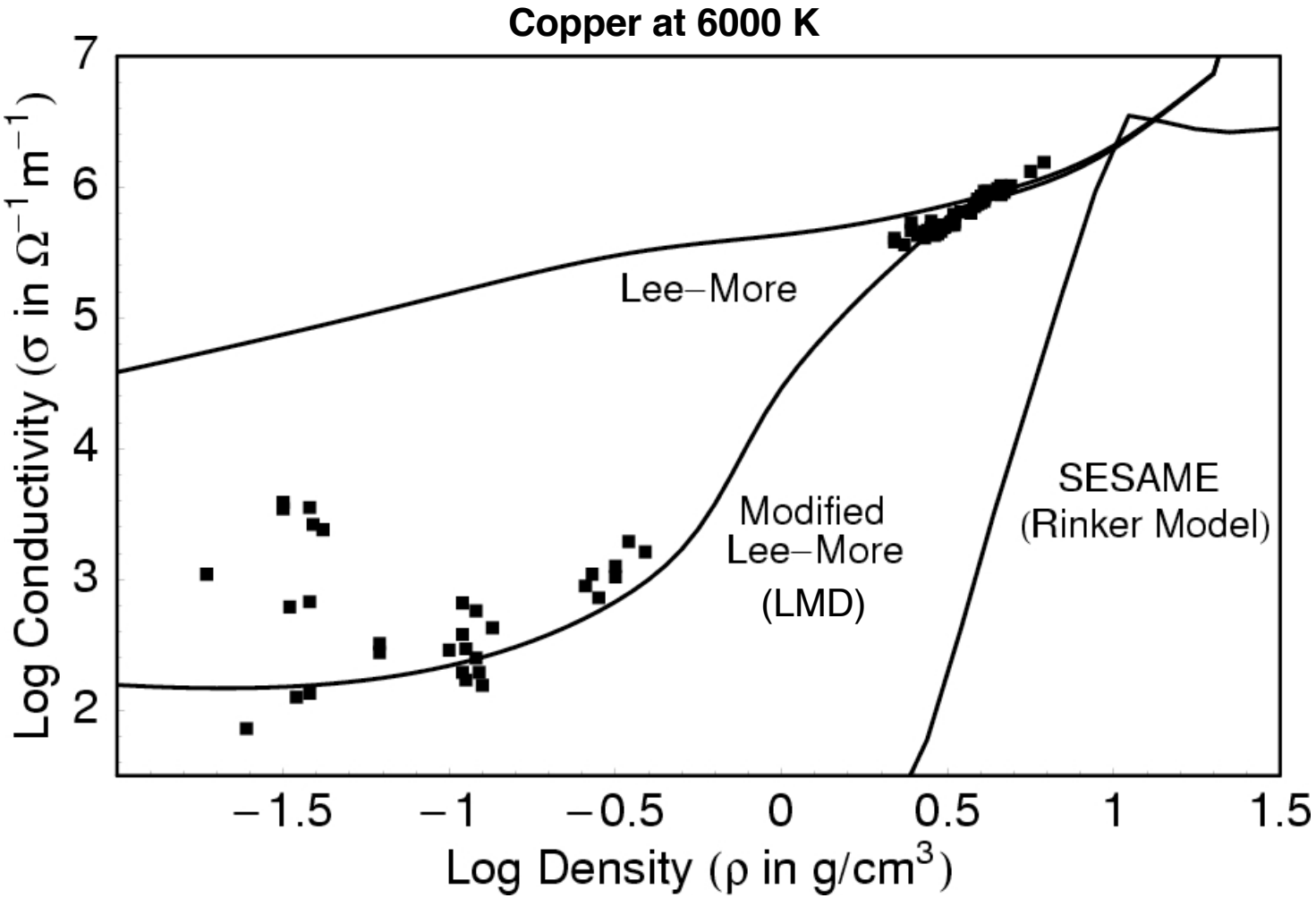
This modified Lee-More model provides much better agreement with DeSilva's copper data



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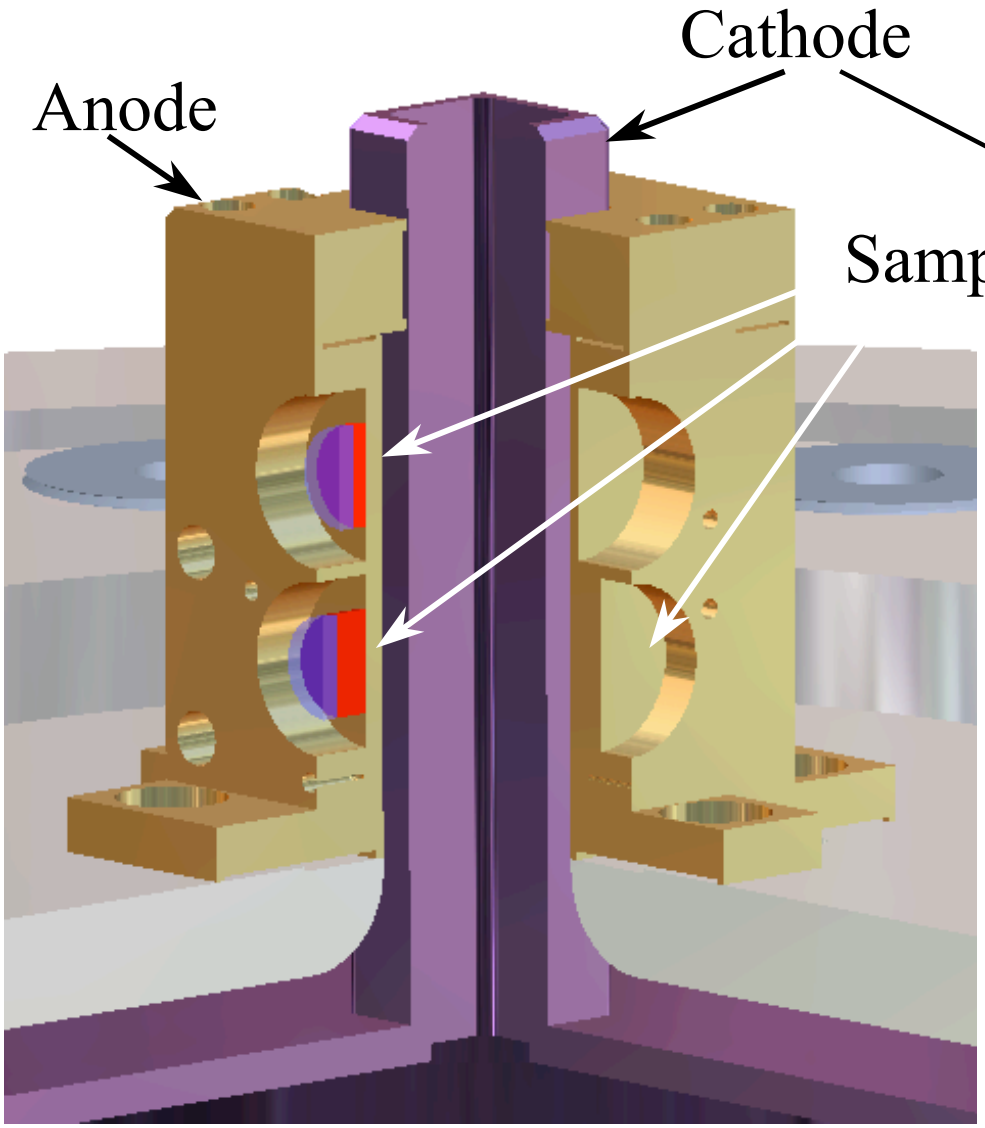
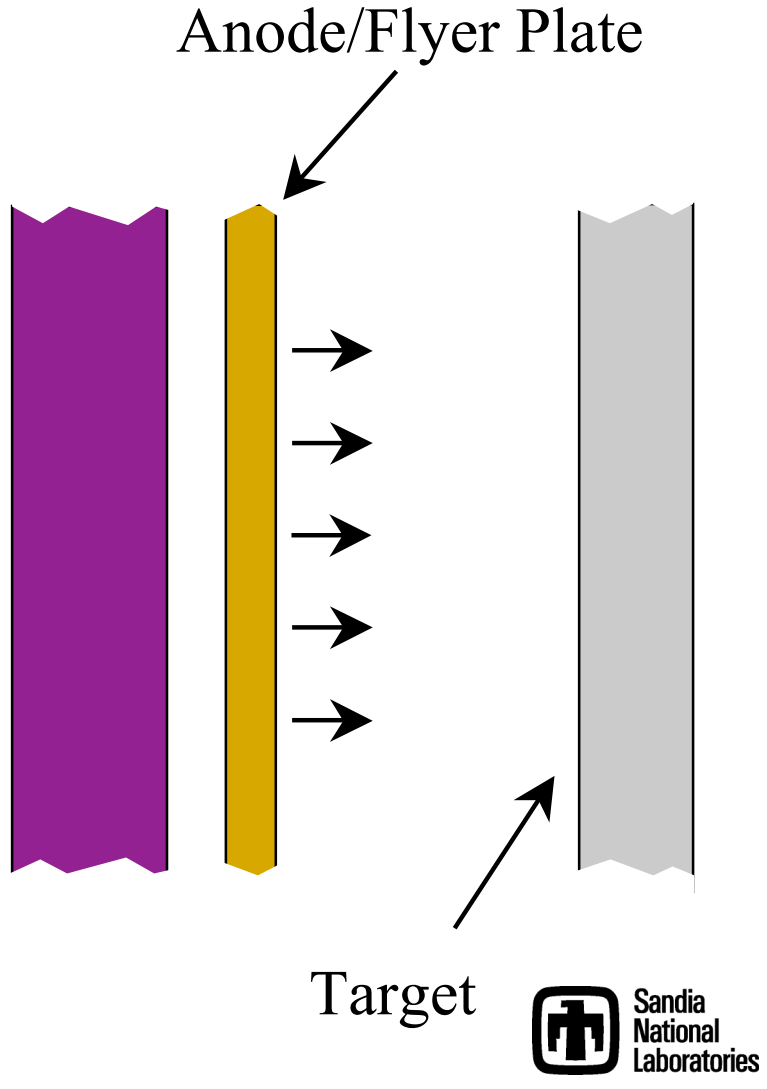
A demanding application: Ultra-high velocity magnetically launched flyer plates (30 km/sec, Multi-Mbar pressures)





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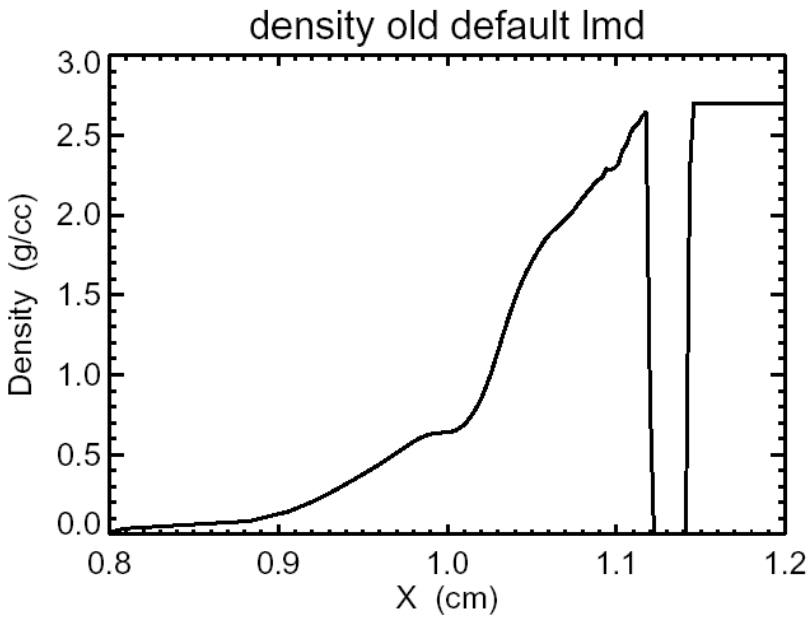
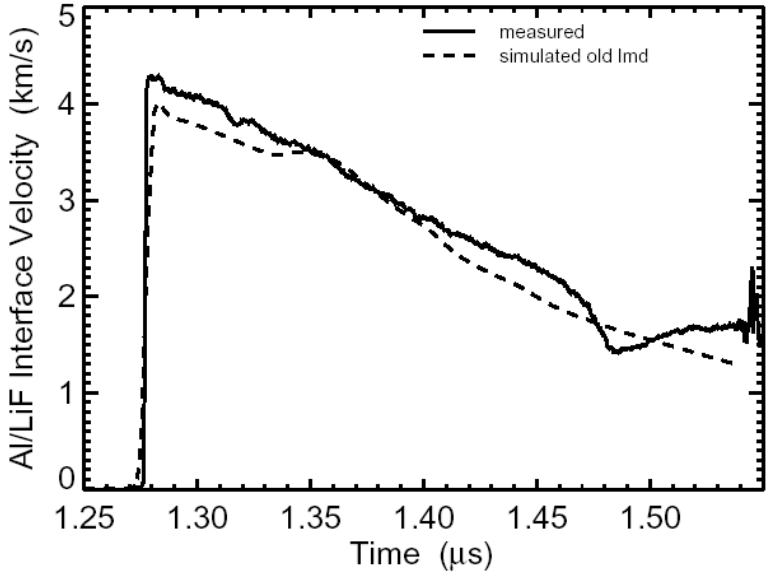
We simulated these magnetically launched flyer plates using the modified Lee-More (LMD) conductivities

Detailed comparison between simulations and experiments for magnetically launched flyer plates suggested that our *improved* conductivities were still not sufficiently accurate for the warm dense liquid aluminum.

Simulations by Ray Lemke with Sandia's 3D Rad-MHD ALEGRA code





For many of our applications we require conductivities accurate to well within a factor of two.

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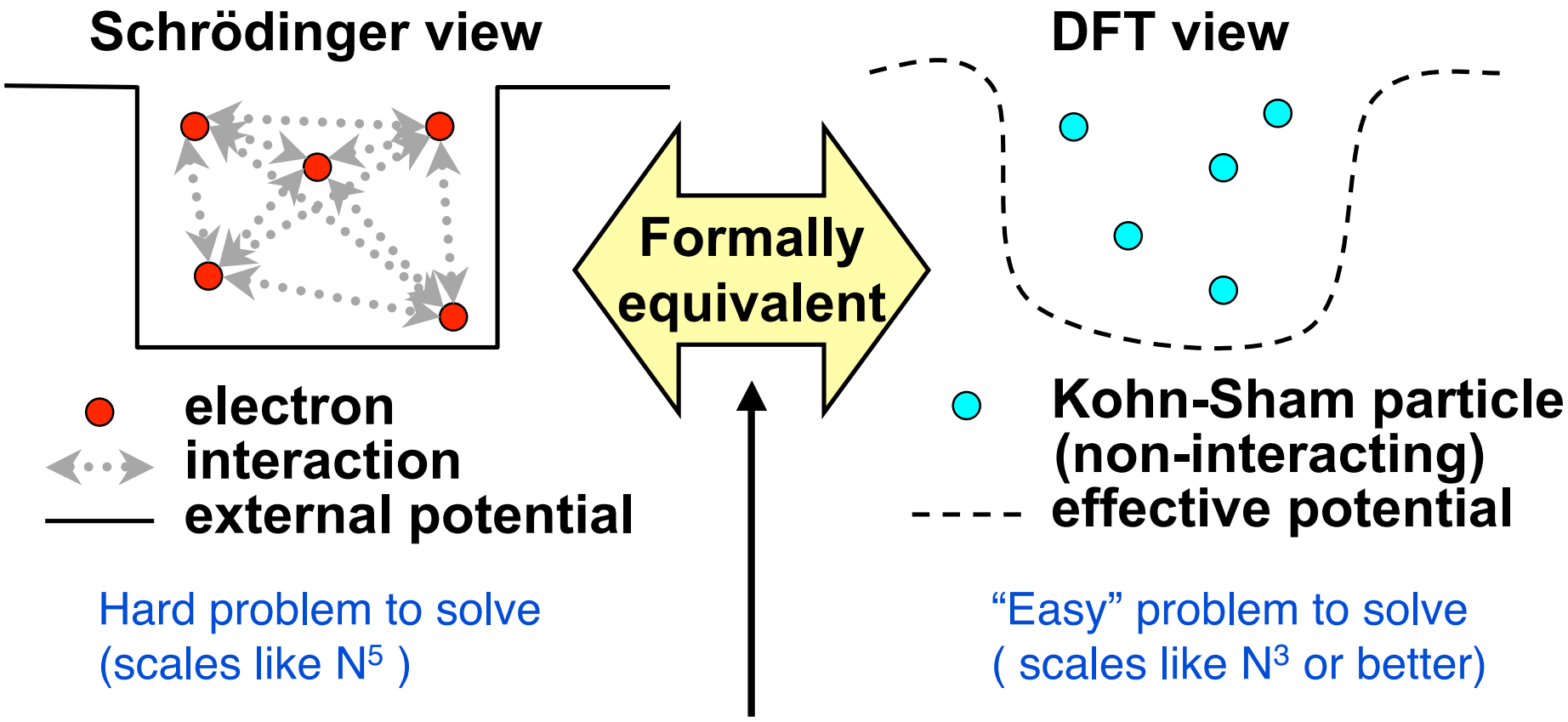
Density Functional Theory (DFT) is a formally exact representation of the N electron Schrödinger Equation



Hohenberg and Kohn proved this (1964)

Nobel prize in Chemistry for Kohn in 1998

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We are using Density Functional Theory (DFT) to perform Quantum Molecular Dynamics (QMD) simulations of Warm Dense Matter

Density Functional Theory is formally exact, but is in practice an accurate approximate solution to the N electron Schrödinger equations. DFT is a work-horse tool in condensed matter physics, but a relative newcomer to high energy density physics.

In addition to our Sandia effort, other groups with a large effort in this area include LANL (Collins, Kress); CEA (Clerouin, Mazevet, Recoules, Blottiau); LLNL (Galli, Gygi, Schwegler)

QMD: The **Kohn-Sham* DFT** equations are solved for a given atomic configuration (fixed in the Born-Oppenheimer approximation) and the **quantum mechanical forces** on all the atoms are calculated from the wavefunctions following the Feynman-Hellmann theorem, **the atomic positions are advanced classically,** and a new solution to the DFT equations is calculated.

*Kohn and Sham,1965

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The Kubo-Greenwood formula is used to calculate the frequency dependent electrical conductivity

$$\sigma_{\mathbf{k}}(\omega) = \frac{2\pi e^2 \hbar^2}{3m^2 \omega \Omega} \sum_{\alpha=1}^{3} \sum_{j=1}^{N} \sum_{i=1}^{N} (F(\varepsilon_{i,\mathbf{k}}) - F(\varepsilon_{j,\mathbf{k}})) \left| \left\langle \Psi_{j,\mathbf{k}} \left| \nabla_\alpha \right| \Psi_{i,\mathbf{k}} \right\rangle \right|^2 \delta(\varepsilon_{j,\mathbf{k}} - \varepsilon_{i,\mathbf{k}} - \hbar\omega),$$

where *e* and *m* are the electron charge and mass. The *i* and *j* summations are over the *N* discrete bands of the triply periodic calculation for the cubic supercell with volume Ω. The coordinate index is α and in general we average over α to improve the statistics. $F(\varepsilon_{i,\mathbf{k}})$ is the Fermi weight corresponding to the energy for the *I*-th band at ***k*** with wavefunction $\Psi_{i,\mathbf{k}}$.

We integrate over the Brillouin zone using the method of special **k**-points

$$\sigma(\omega) = \sum_{\mathbf{k}} \sigma_{\mathbf{k}}(\omega) W(\mathbf{k}) \quad ,$$

and average over 10 to 20 configurations selected from the MD run.

This is really nothing more than the quantum analog of the classical current-current correlation function representation of the conductivity

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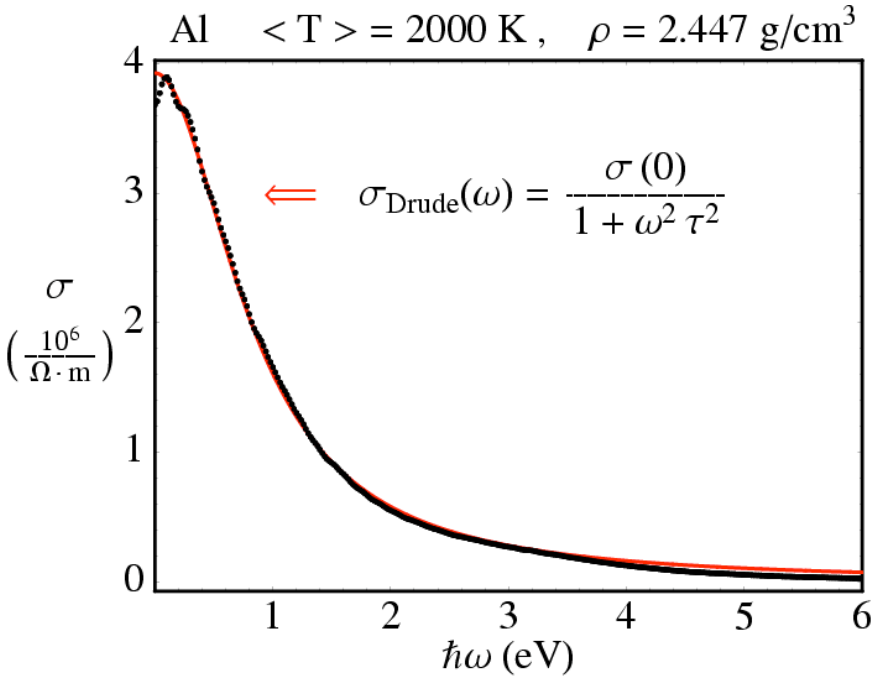
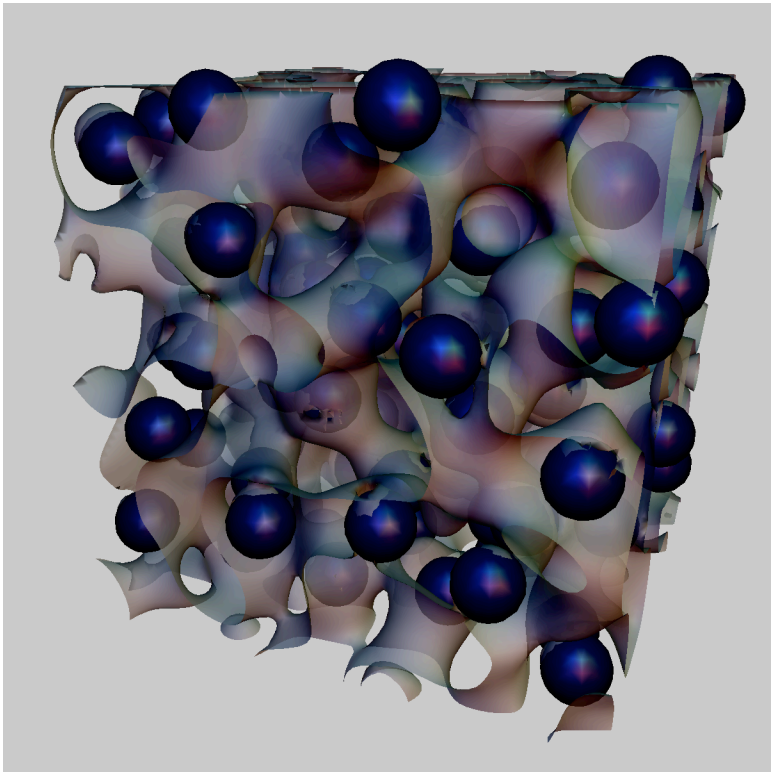
At liquid densities just below solid, the optical conductivity is well fit by the Drude model



The agreement with the Drude model indicates 'nearly free' electrons



Ion cores displayed with iso-surfaces of the mean valence charge density

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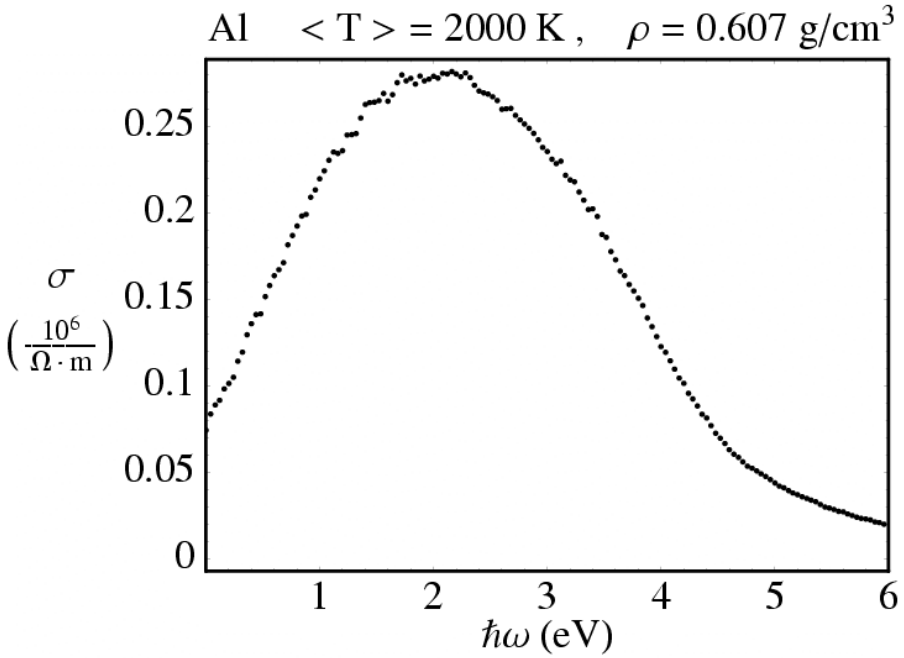
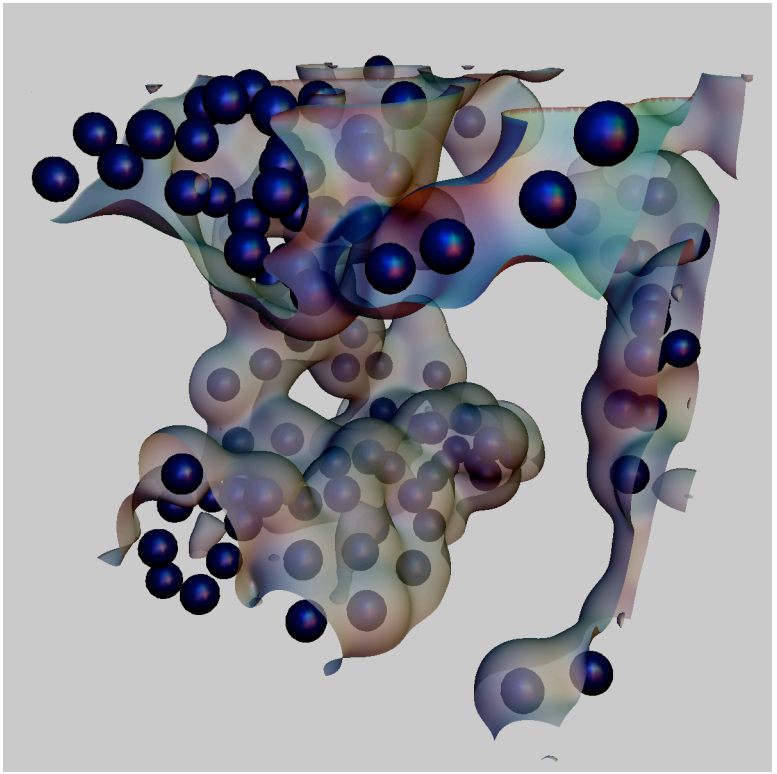
At lower density, where phase separation is pronounced, a gap begins to form at low energy



The dc conductivity has dropped by a factor of 25 for a factor of 4 drop in density



Note the pronounced separation into liquid and void (vapor) regions

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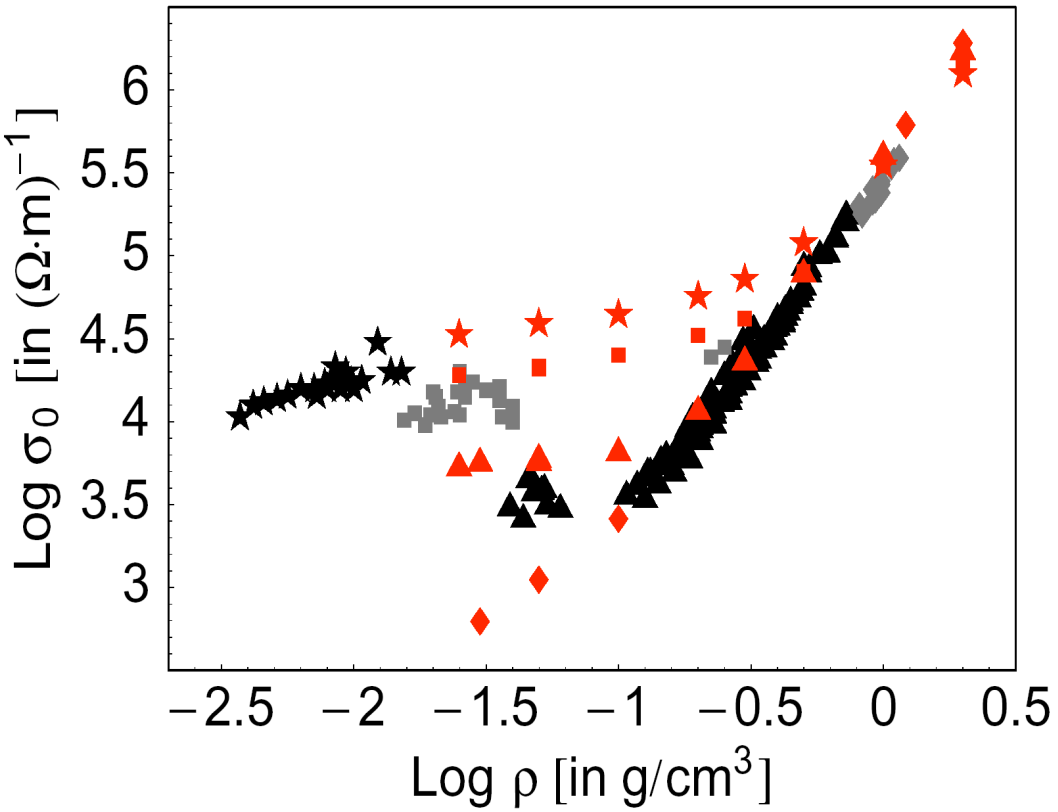
The QMD-KG results are in good agreement with DeSilva's data over a two decade range of density



DeSilva and Katsouros data in black or grey, MD-KG results in red

★ 30000 K, ■ 20000 K, ▲ 10000 K, ◆ 6000 K

[Desjarlais, Kress, and Collins, PRE **66,** 025401(R) (2002)]

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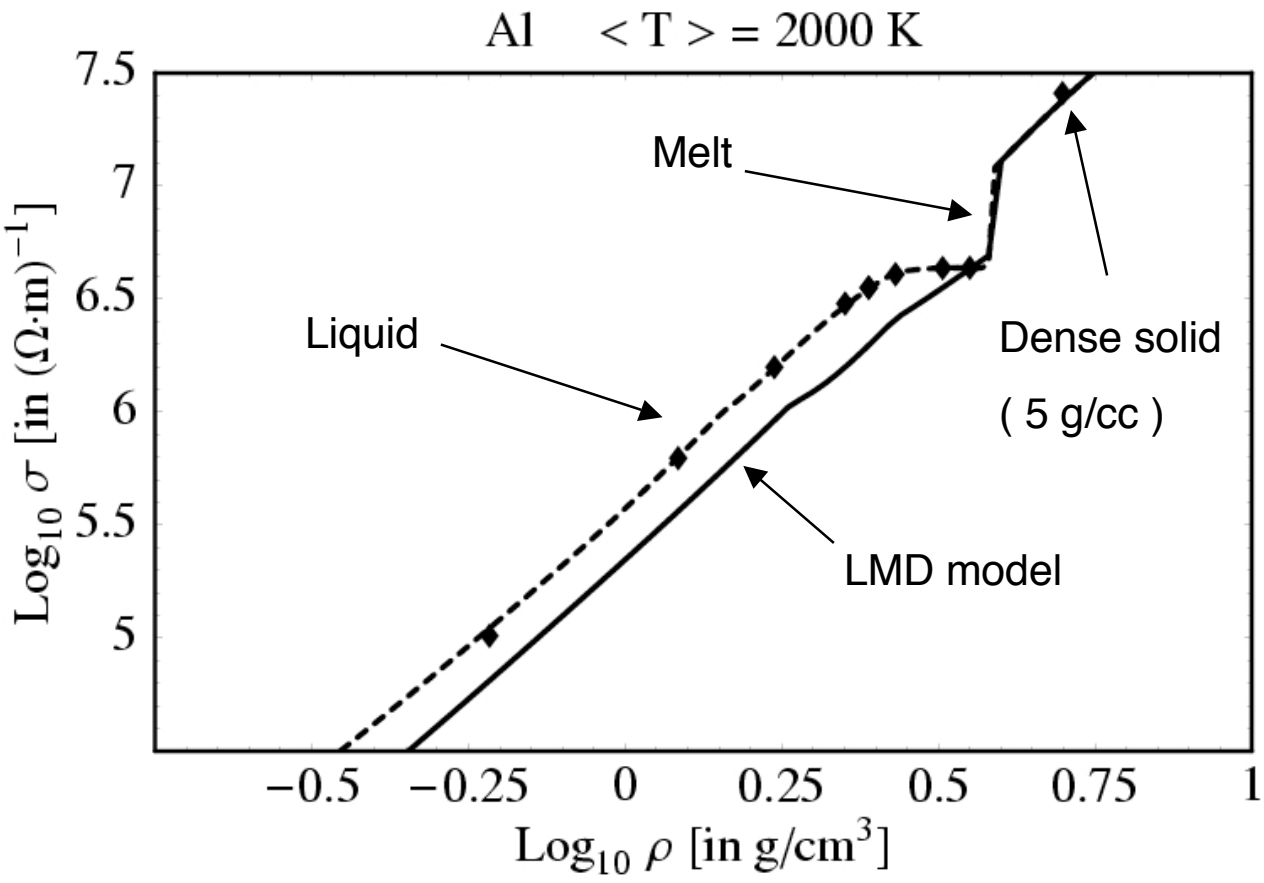
The calculated liquid aluminum conductivities are higher than the *improved* Lee-More (LMD) model predictions



The dashed line shows the 2000 K isotherm from our QMD-tuned wide-range aluminum model

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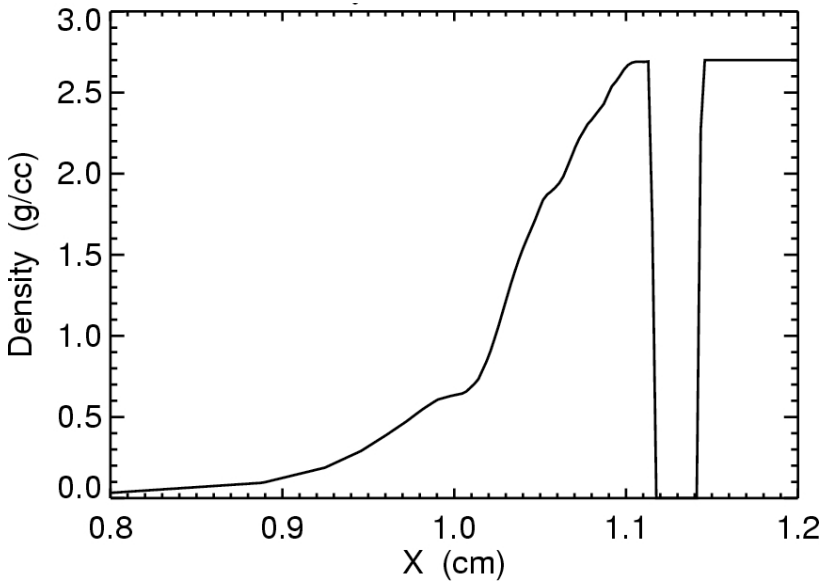
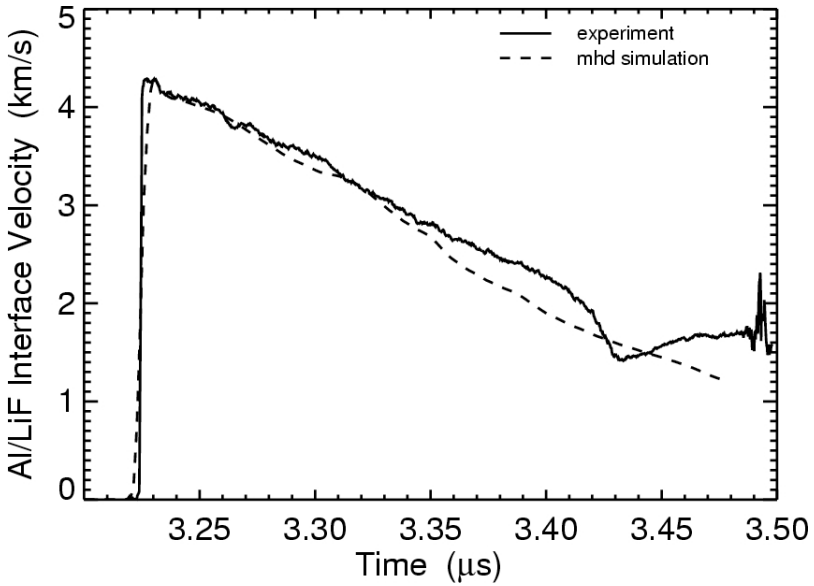
Flyer plate simulations with the QMD based conductivities give very good agreement with experiment

Simulations by Ray Lemke with Sandia's 3D Rad-MHD ALEGRA code





Conductivities based on the QMD calculations have given us a new predictive capability.

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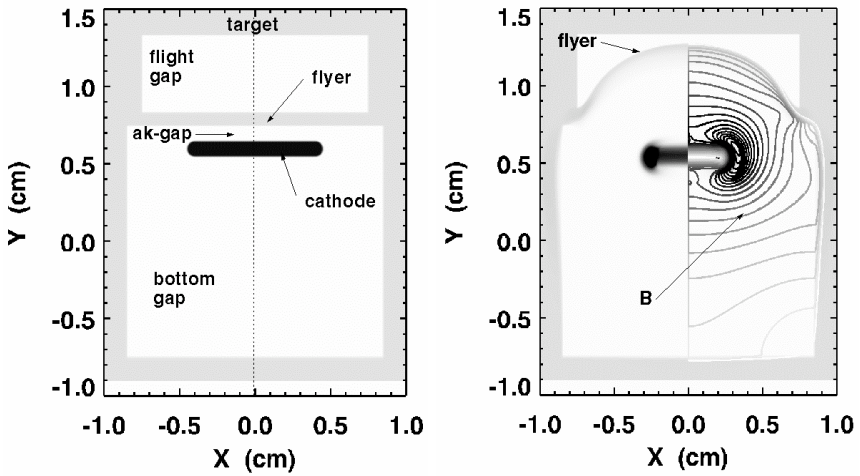
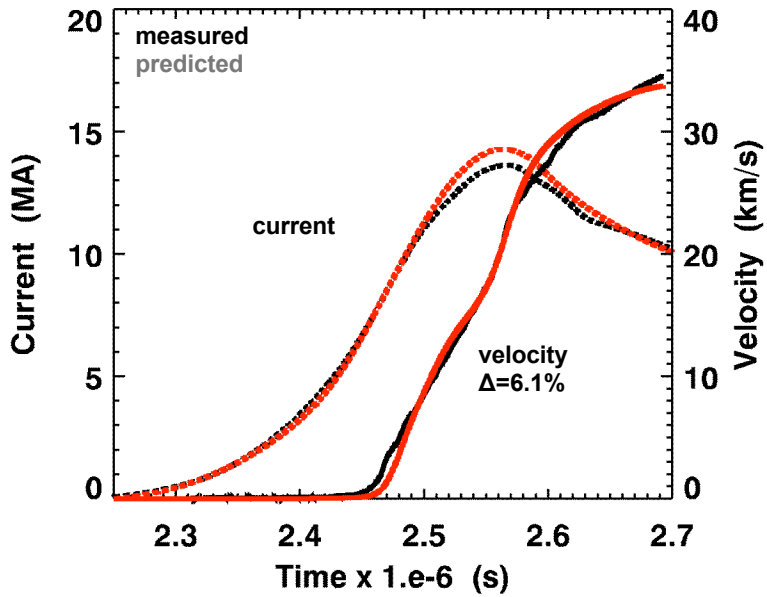
We have used our simulation capability with the new aluminum model to optimize flyer performance on Z



Measured / predicted current & flyer velocity (850 μm Al)



The simulations were performed by Ray Lemke using Sandia's ALEGRA code in 2-D

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Other optical properties are obtained through the Kramers-Krönig relations for $\sigma(\omega)$

Through the Kramers-Kronig relations we have $\sigma_2(\omega) = -\frac{2}{\pi} P \int \frac{\sigma_1(\omega)}{\nu^2 - \omega^2} d\nu$

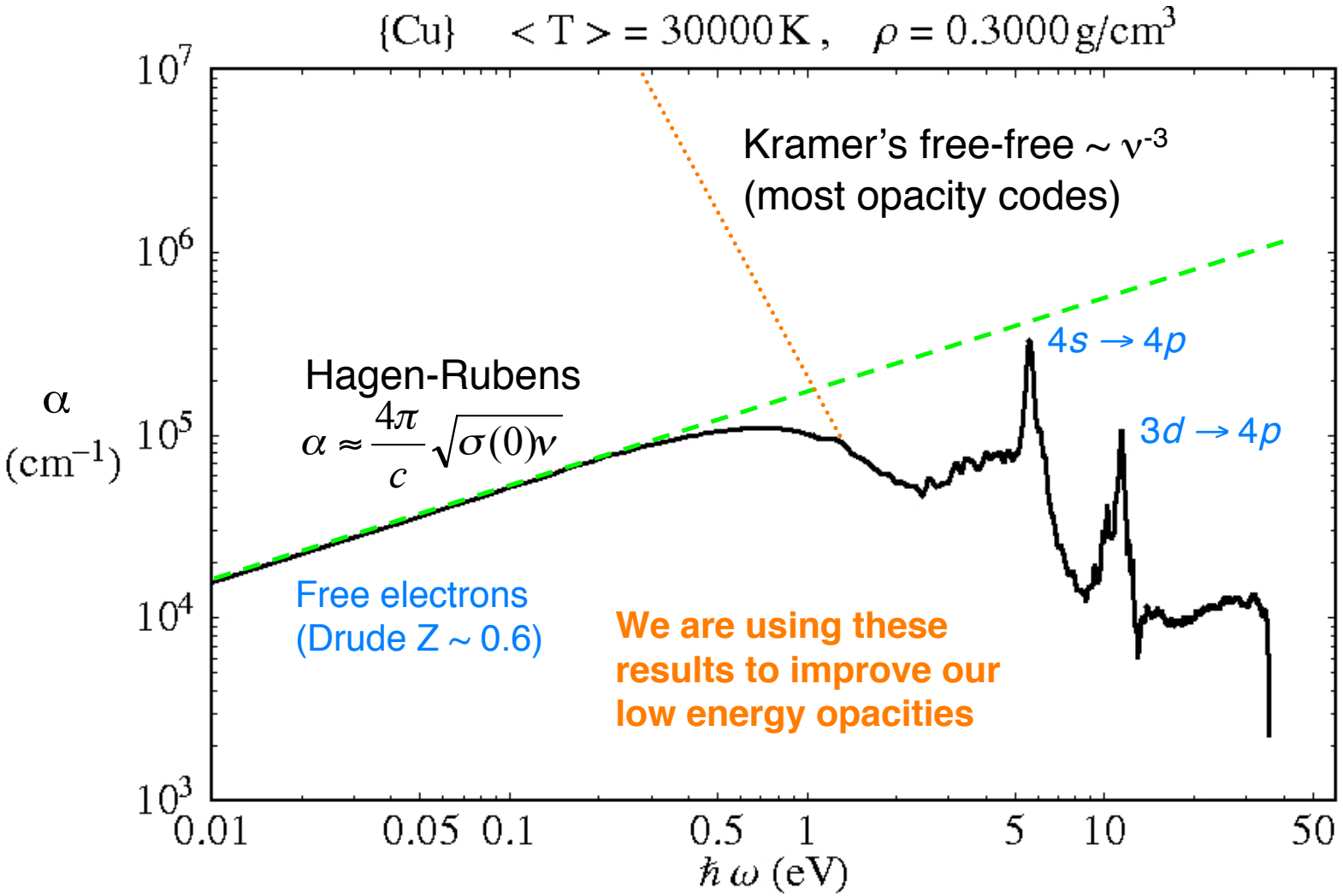
where P indicates the principal value of the integral. The dielectric function is then given by $\varepsilon_1(\omega) = 1 - \frac{4\pi}{\omega}\sigma_2(\omega)$ and $\varepsilon_2(\omega) = \frac{4\pi}{\omega}\sigma_1(\omega)$

Defining $\varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega) \equiv [n(\omega) + ik(\omega)]^2$ we can write the reflectivity $r(\omega)$ and absorption coefficient $\alpha(\omega)$:

$$r(\omega) = \frac{[1 - n(\omega)]^2 + k(\omega)^2}{[1 + n(\omega)]^2 + k(\omega)^2}, \qquad \alpha(\omega) = \frac{4\pi}{n(\omega)c}\sigma_1(\omega).$$

We can now improve our low energy (below ~ 30 eV) opacities $\alpha(\omega)/\rho$ where ρ is the mass density.

The absorption has the proper Hagen-Rubens low frequency behavior, consistent with the dc conductivity



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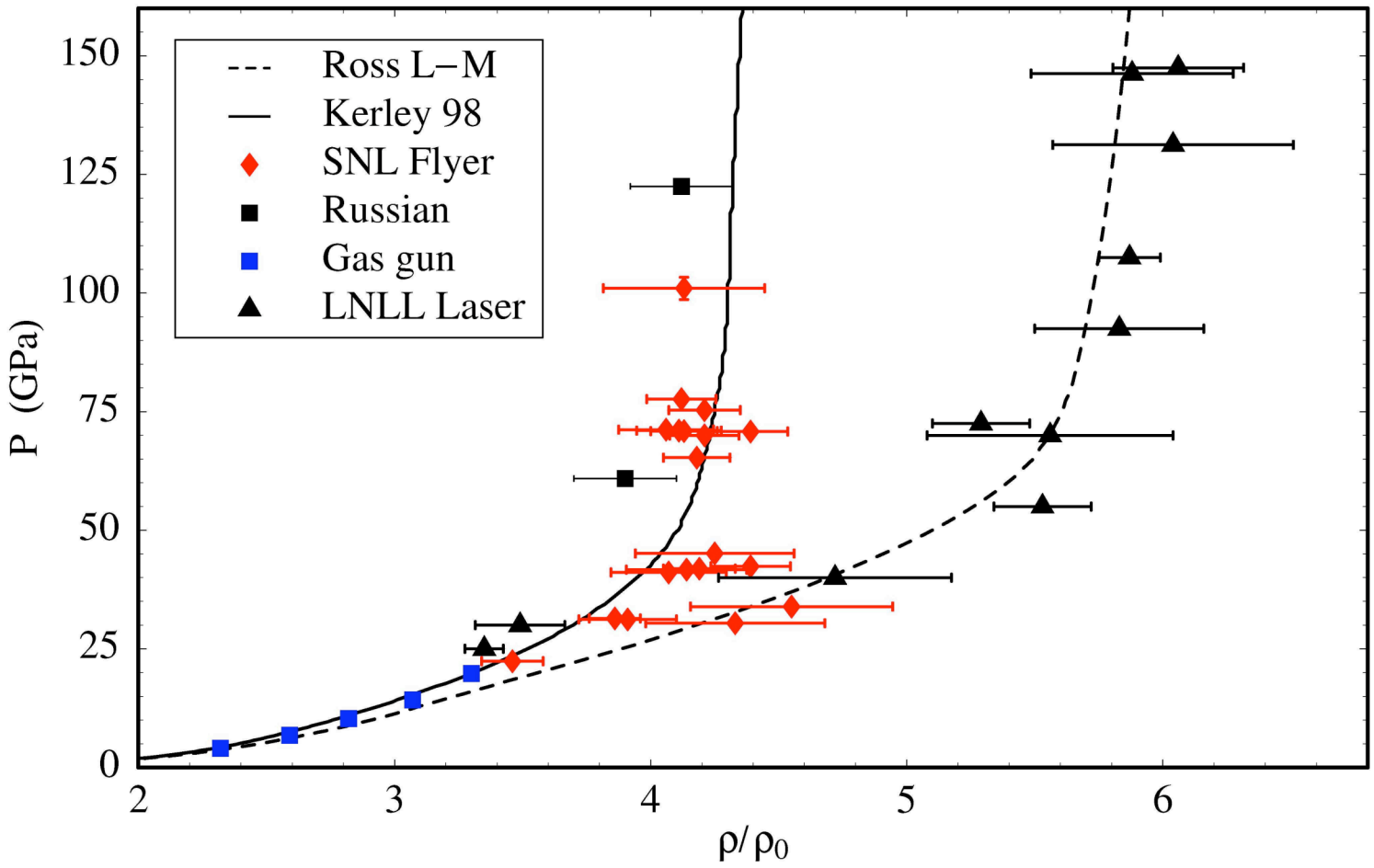
In recent years there has been great interest in the deuterium Hugoniot



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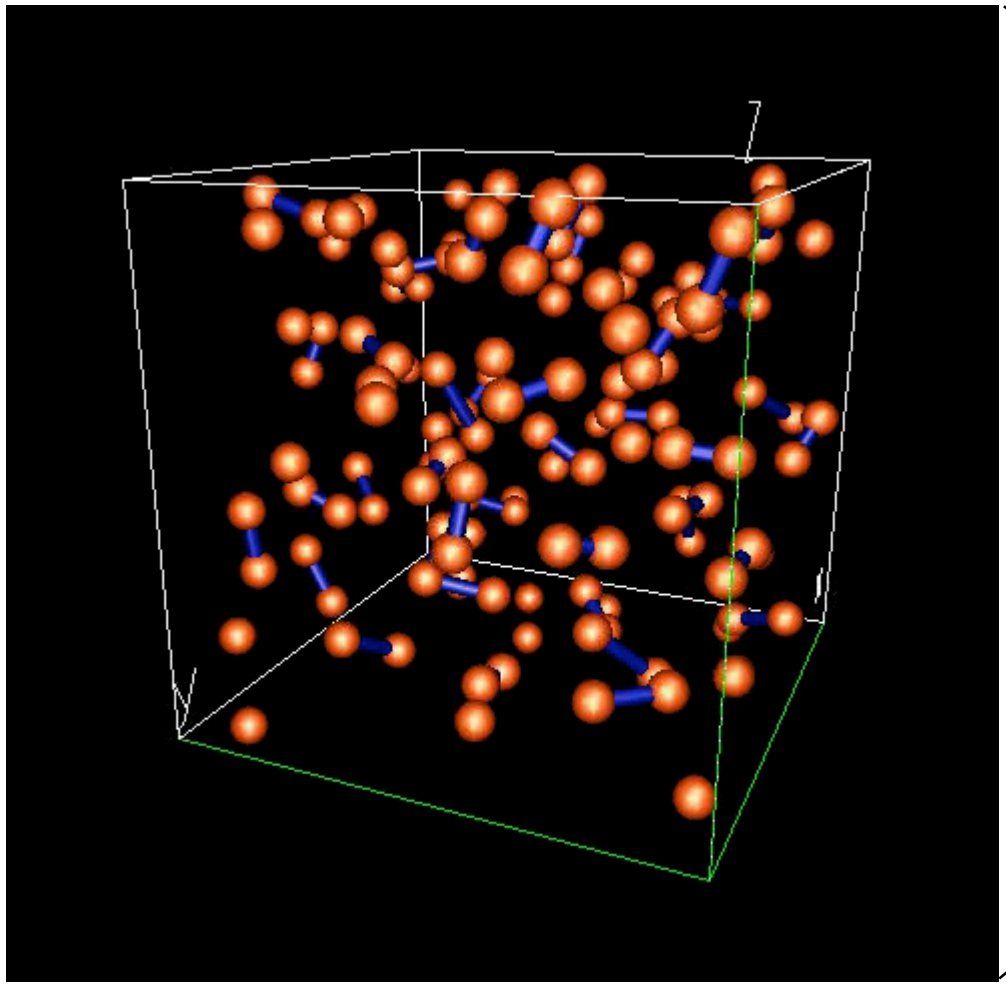
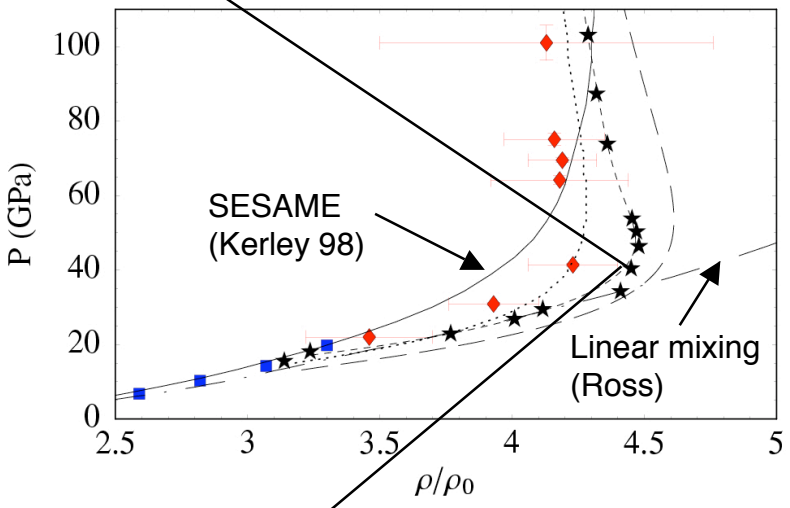
We have used quantum molecular dynamics to study the principal Hugoniot of shocked liquid deuterium



■ LLNL gas gun data (Nellis, Holmes)

◆ Z flyer data (Knudson)

★ QMD



Our QMD simulations of D_2 indicate a Hugoniot in very good agreement with data from Sandia's Z machine

M. P. Desjarlais, Phys. Rev. B **68**, 064204 (2003)

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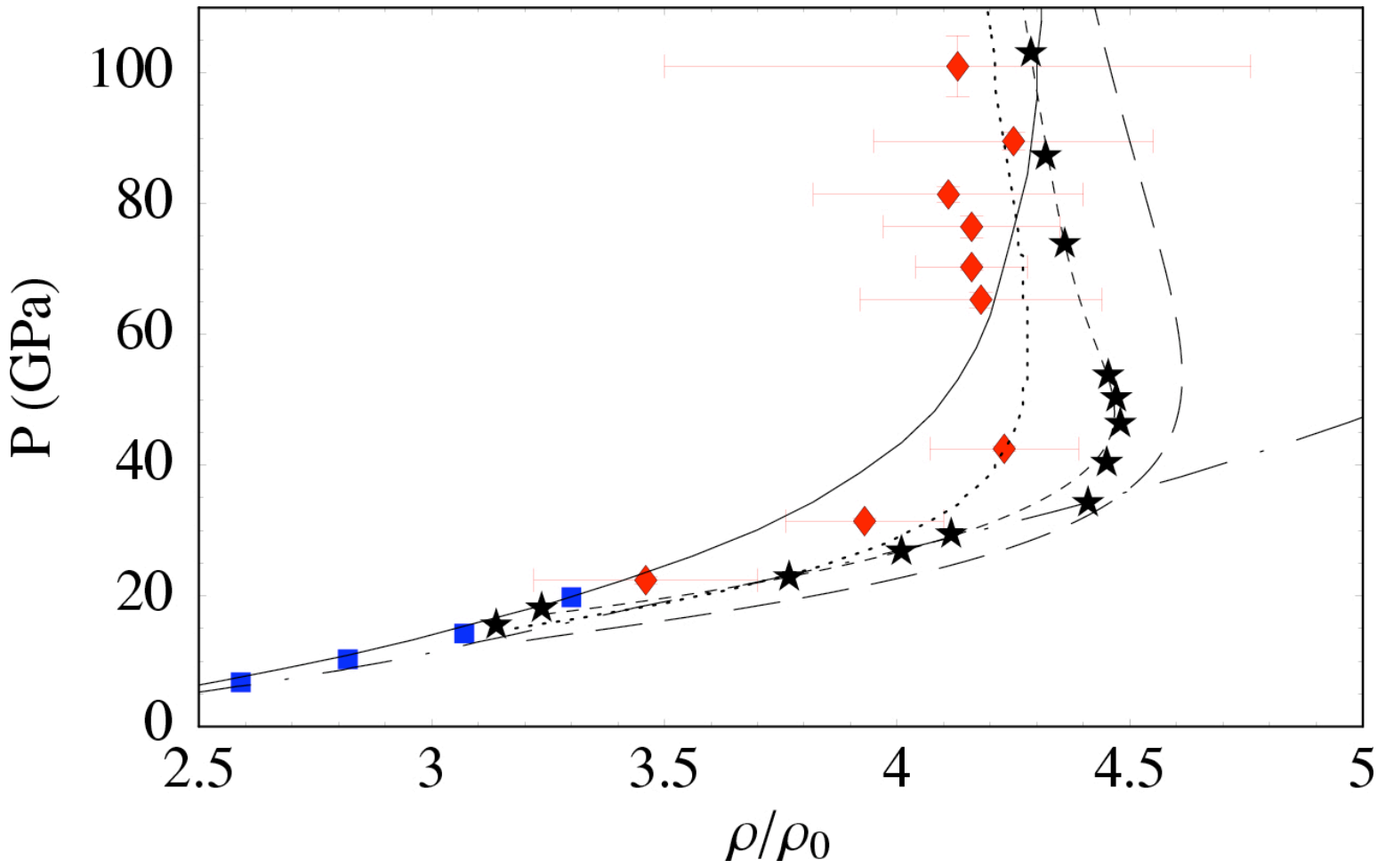
In contrast to earlier DFT/GGA results, our calculations (★) are consistent with the gas-gun data (■)



Temperatures are also in very good agreement with the gas-gun data

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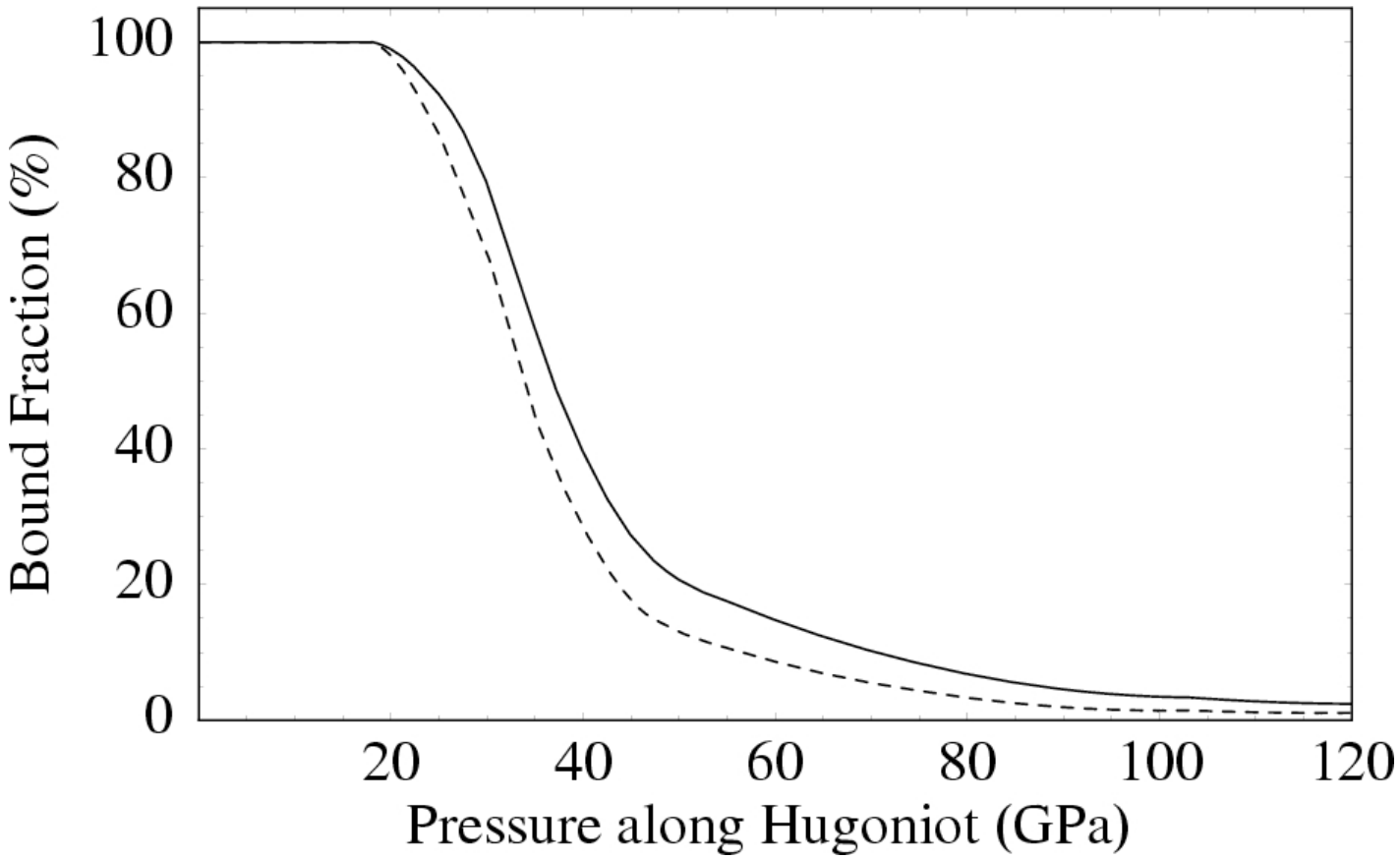
The dissociation along the Hugoniot begins at 20 GPa and is largely over by 50 GPa



Bonds defined for nearest neighbors or mutual nearest neighbors persisting two vibron periods or greater

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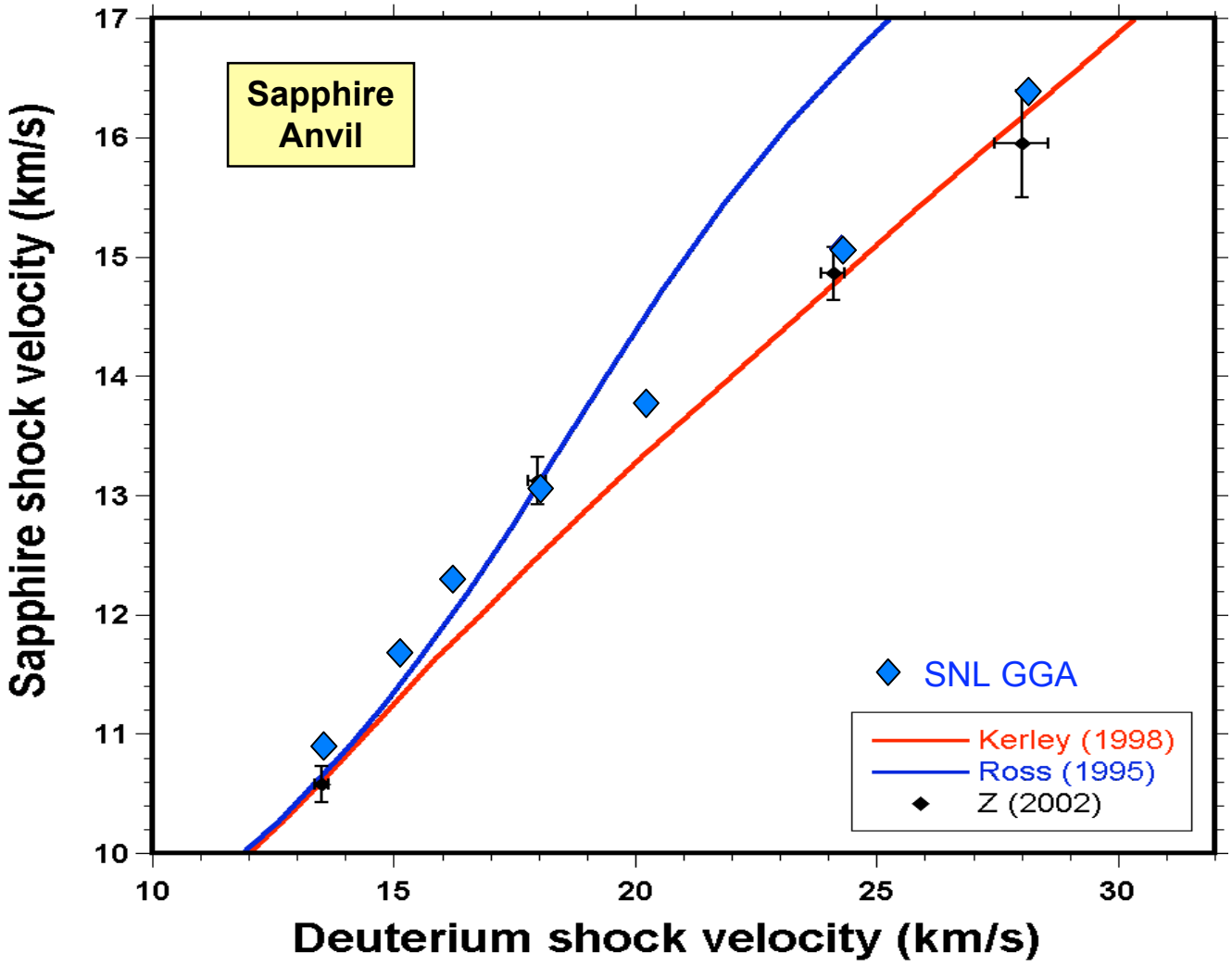
Reshock states with a sapphire anvil are in good agreement with the flyer data from Z



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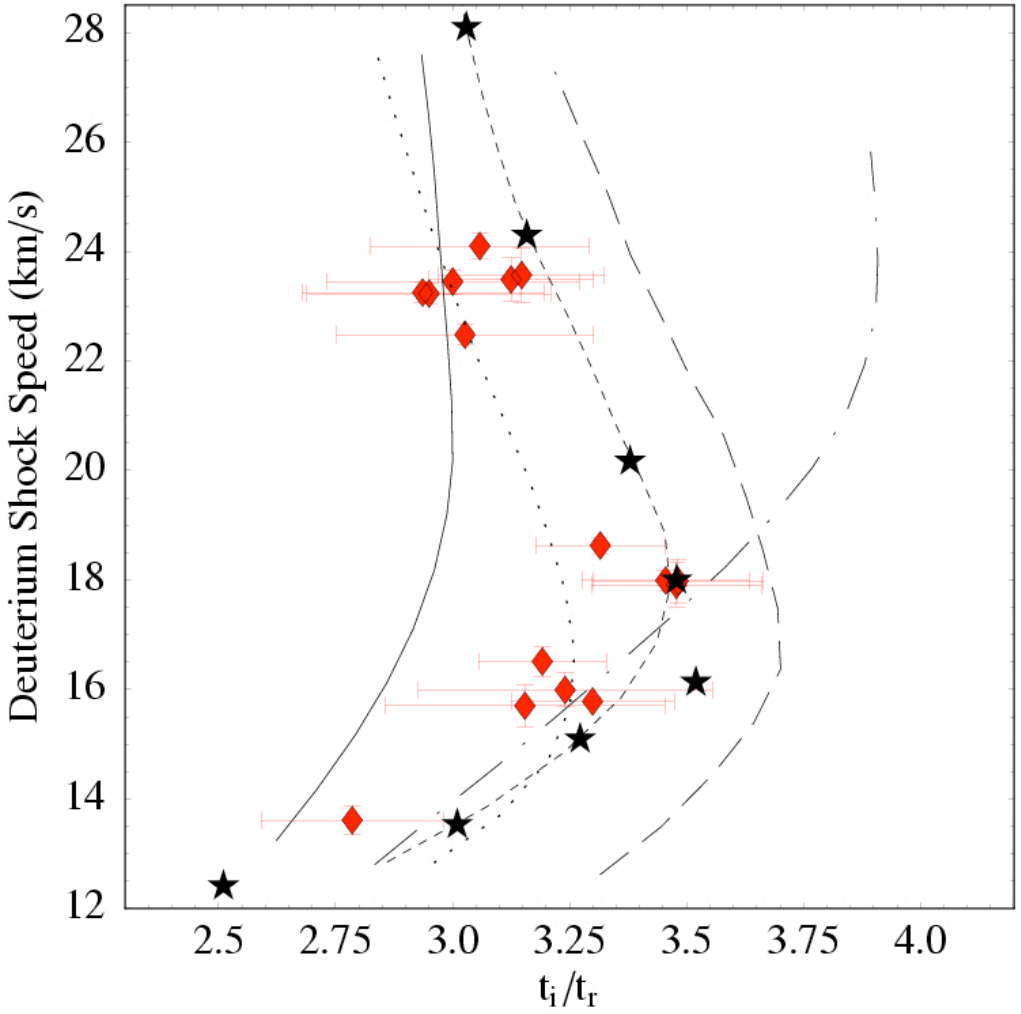
The calculated reverberation timing ratios are in very good agreement with the Z experiments



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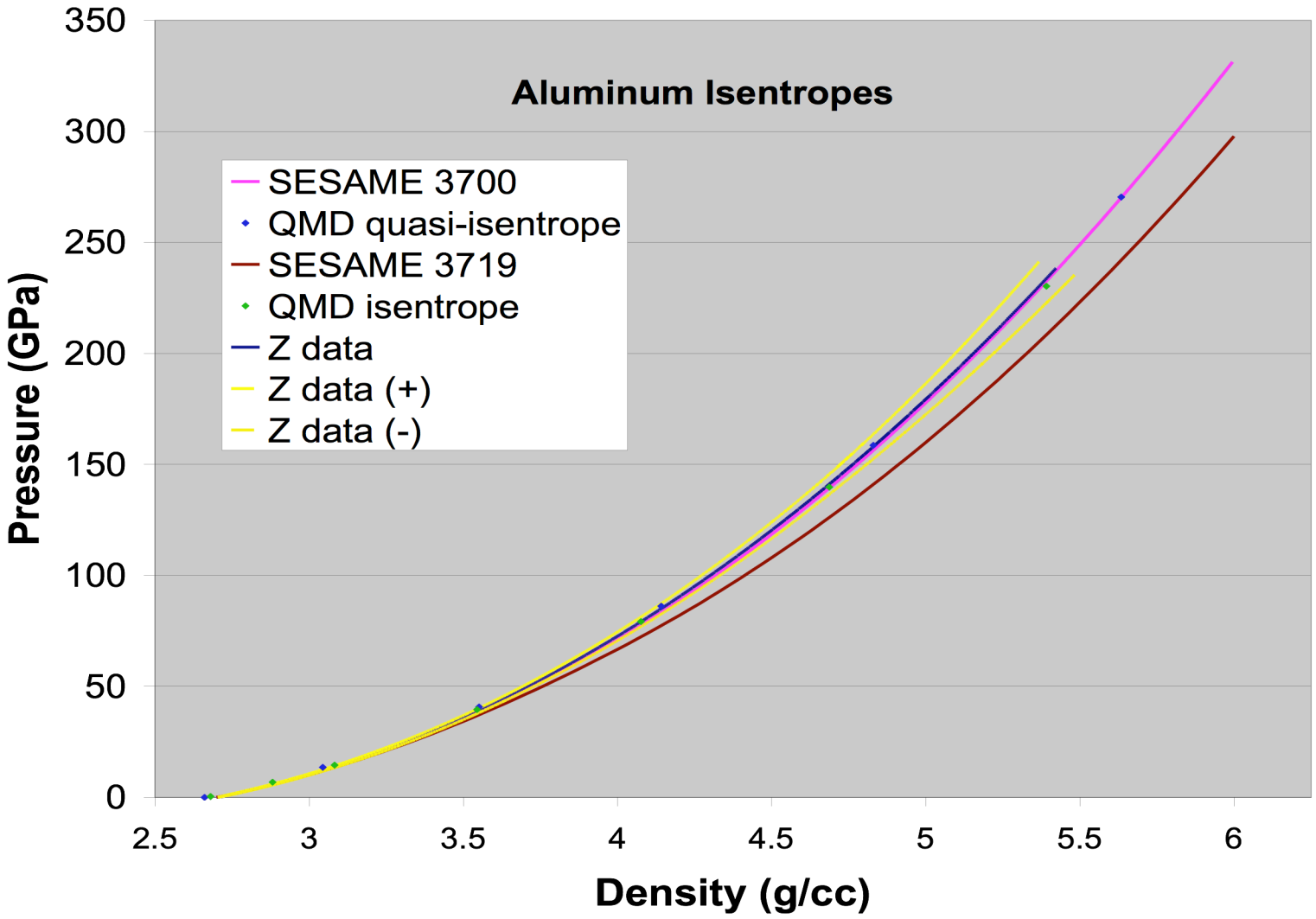
Our calculated principal isentrope for aluminum is in excellent agreement with data from Sandia's Isentropic Compression Experiments (ICE) on Z



Data from **ICE** experiments on Z courtesy of Jean-Paul Davis

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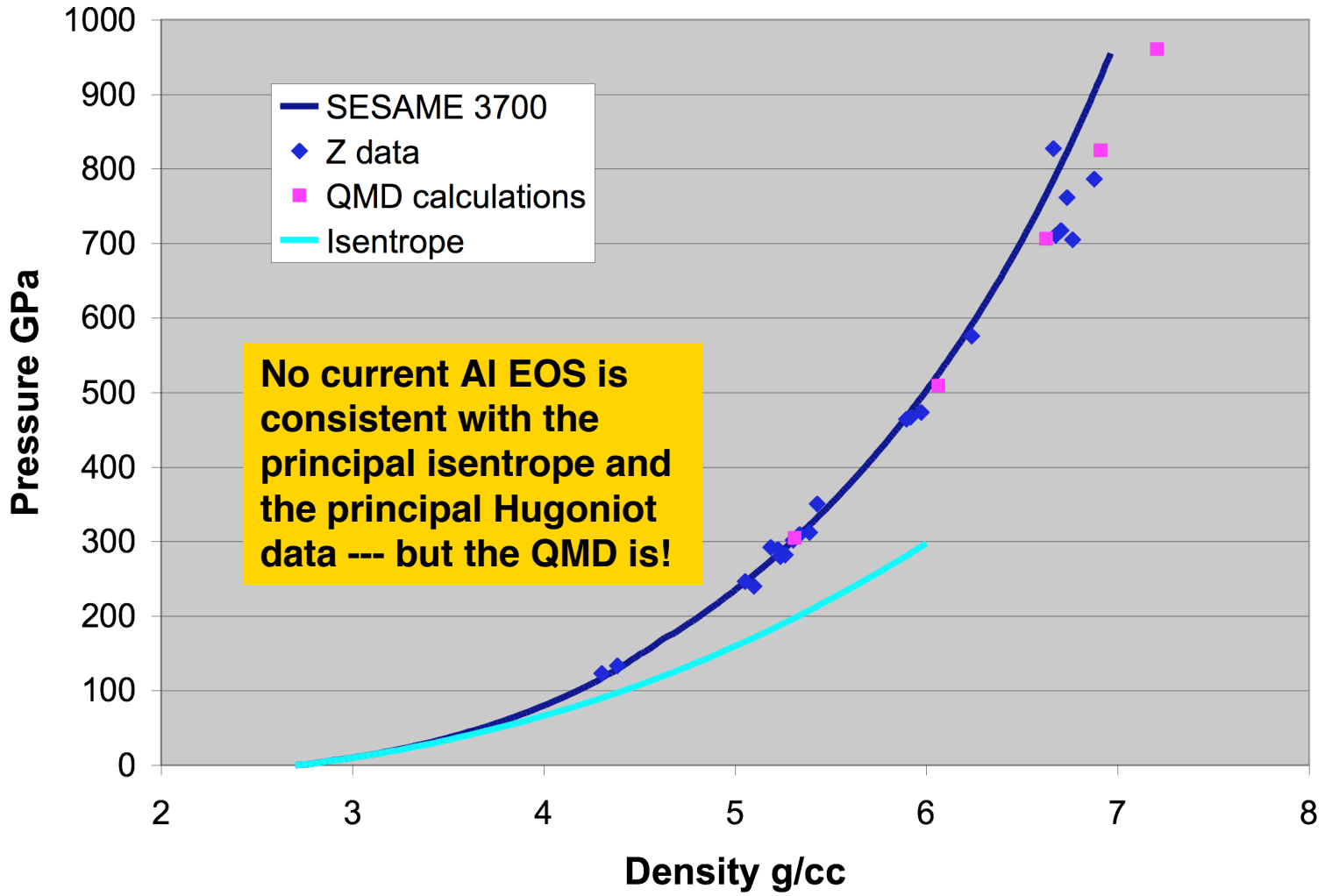
We have also computed the aluminum shock Hugoniot and compared with shock data from Z



Al Hugoniot data from Z courtesy of Marcus Knudson

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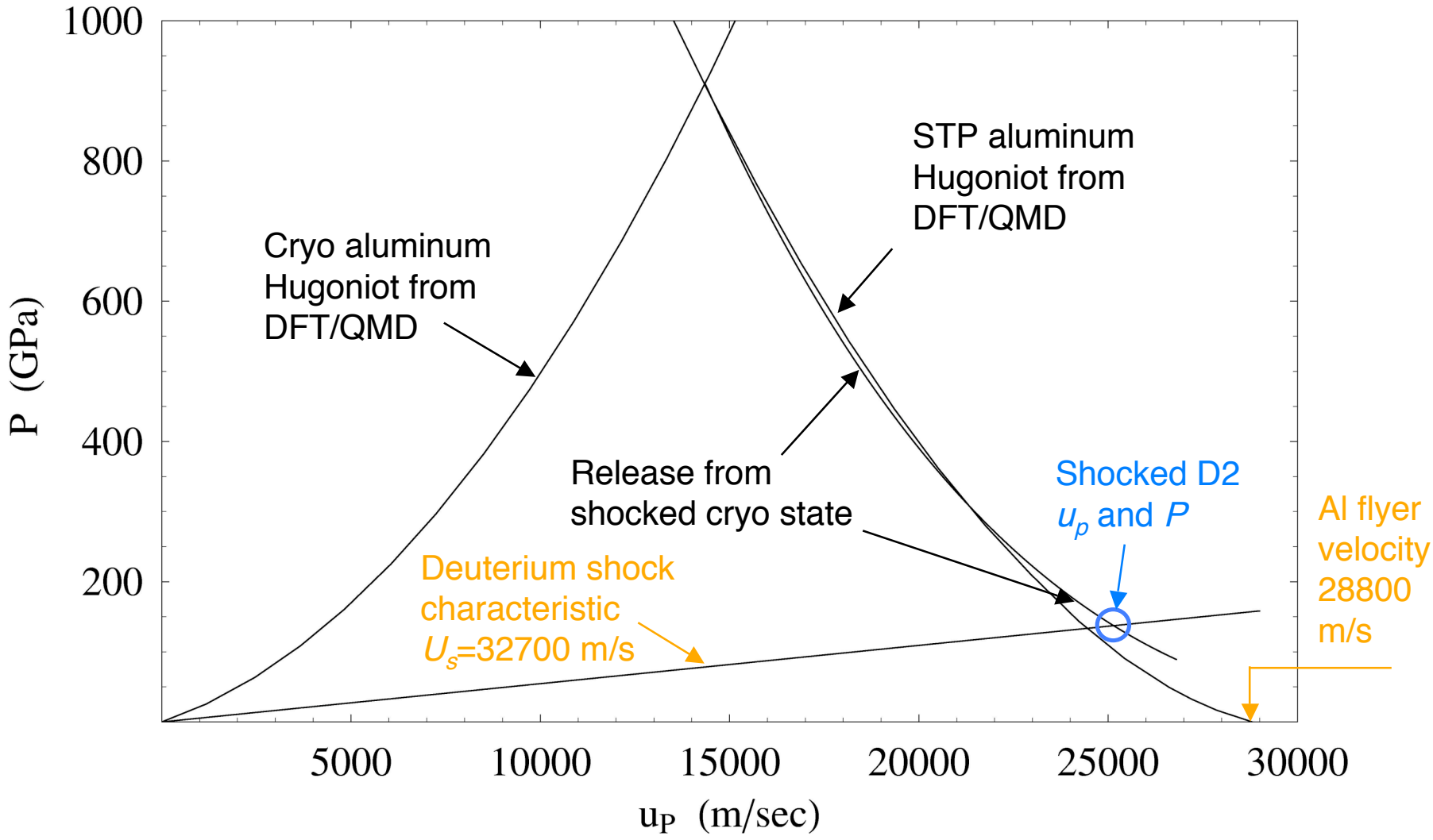
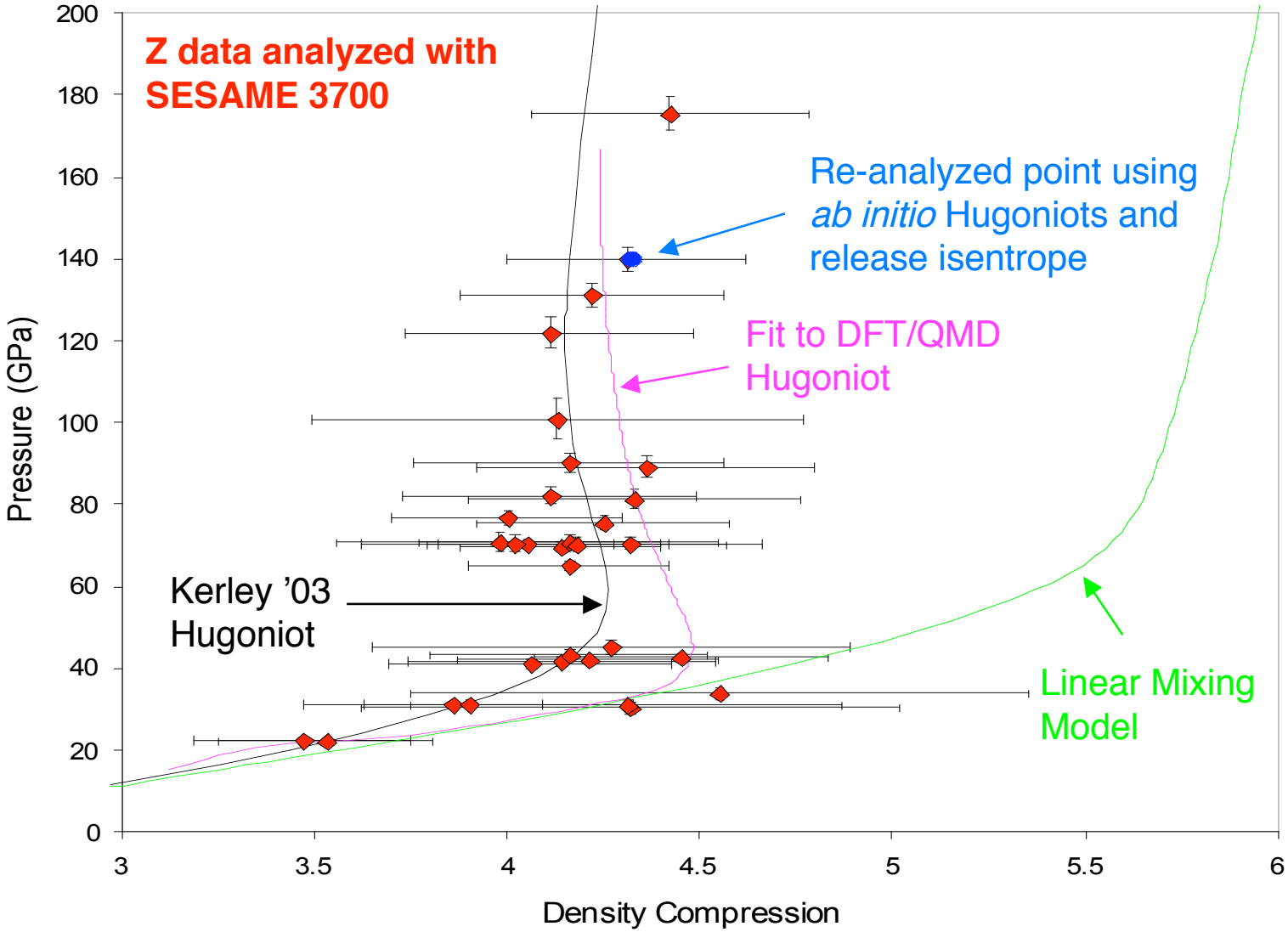
We have all the pieces to do *ab initio* impedance matching calculations



Our re-calculated impedance match point is very close to that obtained with SESAME 3700



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Recent and active research areas

- Principal Hugoniot and reshock properties of deuterium
- QMD based conductivity models for Al, W, Be, and stainless steel
- Liquid-vapor critical points for W and Al
- Principal and release isentropes for aluminum
- Shock melting of Be and Diamond (NIF Ignition Campaign)
- Electrical and thermodynamic properties of water at high energy densities
- Research on advanced electronic structure methods for High Energy Density Physics (finite temperature Exact Exchange, finite temperature GW)

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