# NEAR-TERM OPPORTUNITIES FOR BEAM-DRIVEN WDM SCIENCE

**Richard M. More**, LBNL

- **Remarkable phenomena in WDM** <-- *What's there in WDM?*
- **Tools and applications** <-- *Sketch of today's research*
- **Basic questions** <-- *There are many questions*

WDM conditions are easily produced in the laboratory

$0.1\ \text{eV} < T < 10\ \text{eV}$ $\qquad$ $0.1\ \rho_0 < \rho_0 < 10\ \rho_0$

***Surprising that many basic science questions are open***

# *WDM Research*

**Warm + dense ==> high pressure ==> dynamic experiments**

"*Warm*" -- > Melt and boil anything, even tungsten

2-phase (liquid-vapor)

Electronic excitation and maybe ionization

"*Dense*" -- > Atoms interact with neighbors (disordered fluid environment)

"*Matter*" -- > Material phases and properties

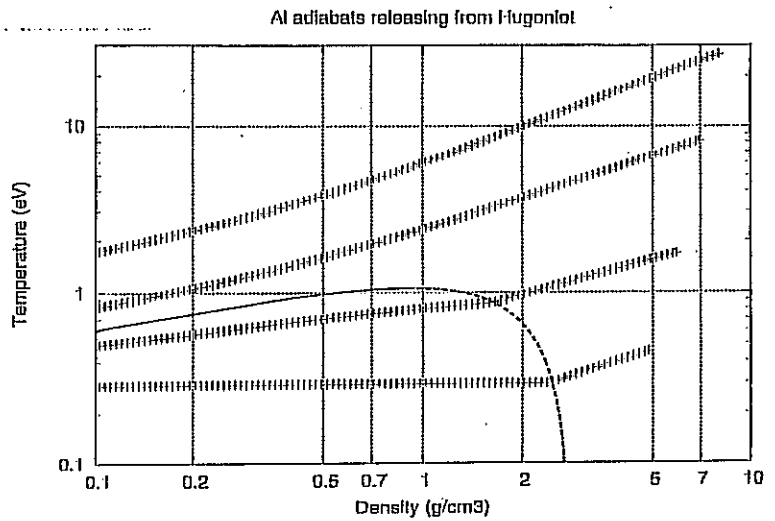
"***Research***" **means we want to *learn something new*.**

# *Typical WDM experiments*

A Heat slowly

B Heat rapidly

C Shock heat & compress




| | |
|---|---|
| Time before disassembly | $\tau \sim L / c_s \sim (1\ \mu) / (10^5\ \text{cm/sec}) \sim 10^{-9}$ sec |
| Specific heat | $C_V \sim 3$ eV/atom-eV $= (2.9/A)\ 10^5$ Joules/gram-eV |
| Energy required | For $\rho \sim 10$ g/cm$^3$ and $T \sim 1$ eV $\quad E = \rho L C_V T \sim 15$ J/cm$^2$ |
| Power | $E / \tau \sim 1.5\ 10^{10}$ W/cm$^2$ |
| | |
| For shock launched by a flier: | Assume a 5 mm flier, $\rho = 10$ g/cm$^3$, $v = 10$ km/sec |
| Energy/area | $E = 1/2\ \rho L v^2 = 2.5\ 10^5$ Joules/cm$^2$ is delivered in 1/2 μsec |
| Power | $5\ 10^{11}$ W/cm$^2$ |

Ticket price

# *Warm Dense Matter is a distinct subject of study*

Existence theorem

USP Laser absorption experiment D. Price et al., PRL 75, 252 (1995)

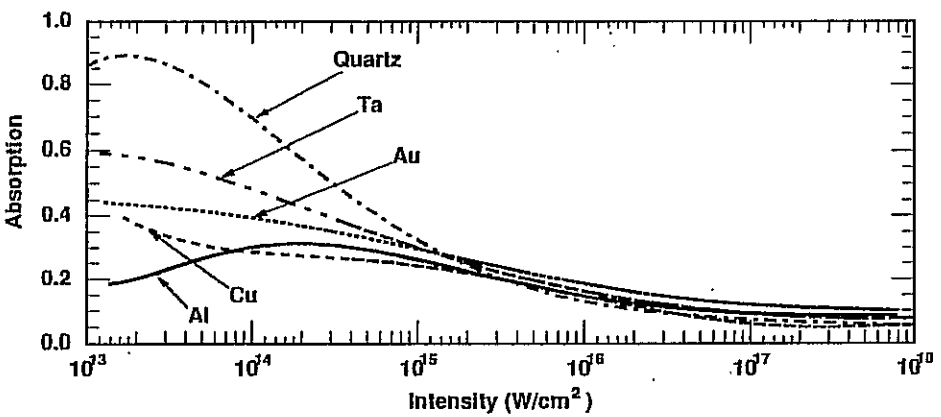
120 fsec pulse - normal incidence - self-absorption $I = 10^{12}$ to $10^{18}$ W/cm$^2$

Aluminum is well-described by "usual" plasma theory (QEOS + Drude)

Other materials agree with Al at high-T (plasma) conditions

**Intermediate range:**

Not = solid
Not = plasma
Not a simple interpolation




***each material is unique***

The experiment shows there are new things to learn.

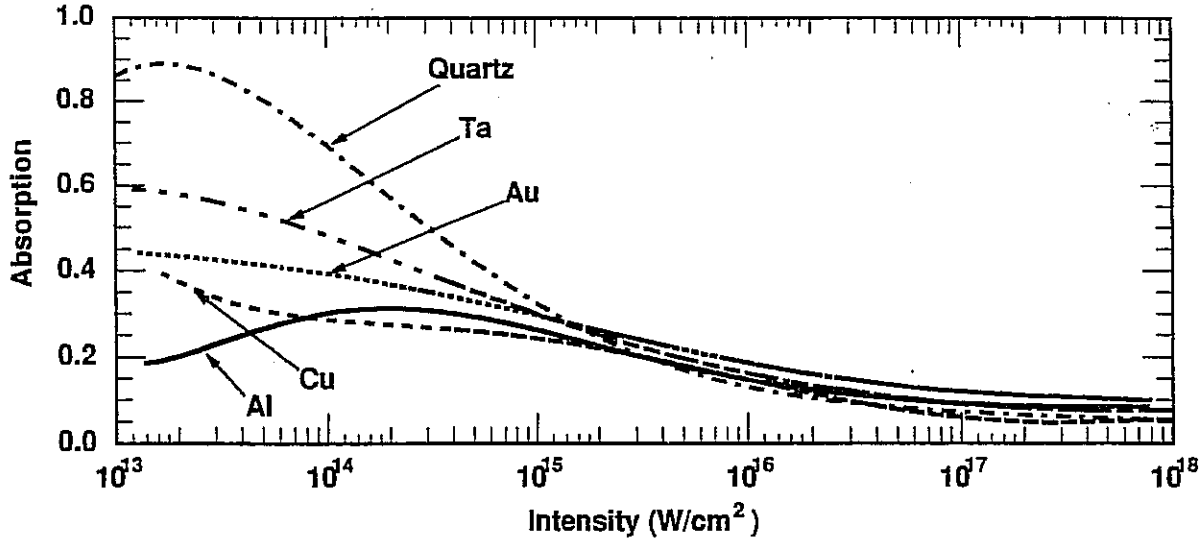



FIG. 1. Absorption fraction vs peak laser intensity for aluminum, copper, gold, tantalum, and quartz targets. In Figs. 1, 3, 4, and 5 laser intensity is the temporal and spatial peak value of the laser intensity.

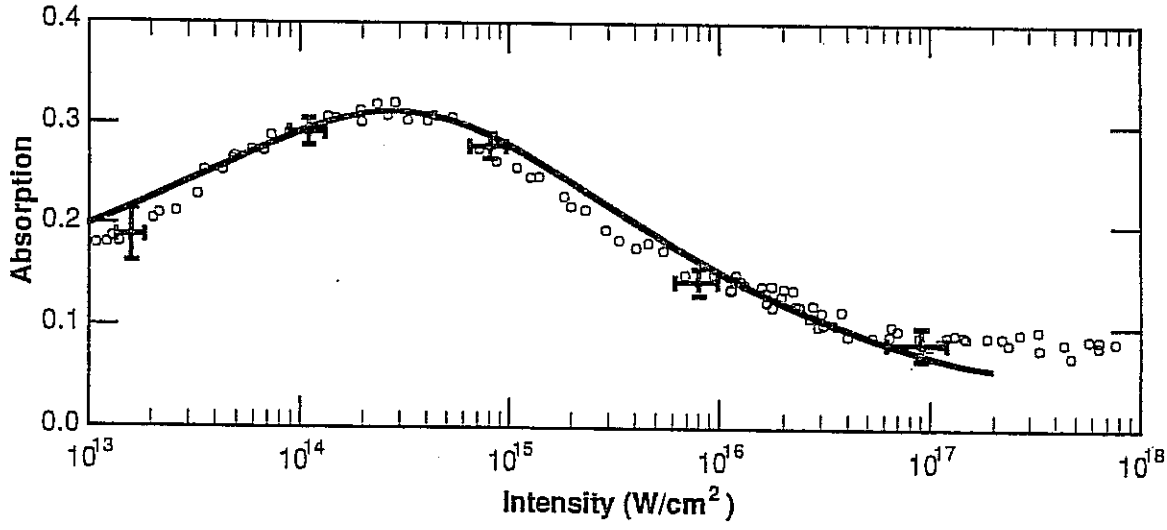



FIG. 5. Measured and calculated absorption fractions for an aluminum target vs peak laser intensity. The calculation assumes a Gaussian laser spatial profile. Error bars shown indicate systematic uncertainty in intensity and random error in absorption. The absolute absorption scale is believed to be known to $\pm 0.035$.

D. F. Price, R. M. More, R. S. Walling, G. Guethlein, R. L. Shepherd, R. E. Stewart and W. E. White,

Physical Review Letters **75**, 252 (1995).

"Absorption of ultra-short laser pulses by solid targets heated rapidly to temperatures 1 - 1000 eV"

*Scientific value of experiments increases with their precision, so good diagnostics are essential.*

$E_{dep}$, expansion L(t), electrical I(t), V(t), optical emission $I_\nu$, XRD

At least four possible methods to measure temperature:

| | | |
|---|---|---|
| Hydrodynamic release | $L \sim 3\, c_s(\rho, T)\, t$ | 3 --> ideal gas |
| Electrical conductivity | $\sigma(\nu, T)$ = current / **E** field | |
| Optical emission | $I_\nu = \varepsilon(\nu, \rho, T)\, B_\nu(T)$ | $\varepsilon \leq 1$, $\varepsilon = a$ |
| X-ray diffraction or scattering (Thomson scattering) | | |

Need to cross-calibrate temperature scales.

**Fixed points** will help for this.

**phenomena that occur in a narrow range of temperatures.**

# *Remarkable phenomena can be predicted*

Predictions from preliminary theory and experimental clues

- o $\pm$ ion plasma [ $n_e << N_-$ ]
- o Metal-Insulator transition [ high $\sigma$ --> low $\sigma$ ]
- o "Black glass" [ $Im(\varepsilon) > Re(\varepsilon) > 0$ ]
- o Neutral phase of ionic solids ?
- o Mixed valence and shell-crossing [e.g., $Pm^{9+}$]

Sharply-defined observable phenomenon can help calibrate temperature scales

# PLUS-MINUS ION PLASMA

Surprising electronegative plasma predicted for WDM temperature range.

plus and minus ions  but  $n_e << N_+$

Charge density $10^{17}$ $cm^{-3}$ like semiconductor carrier density

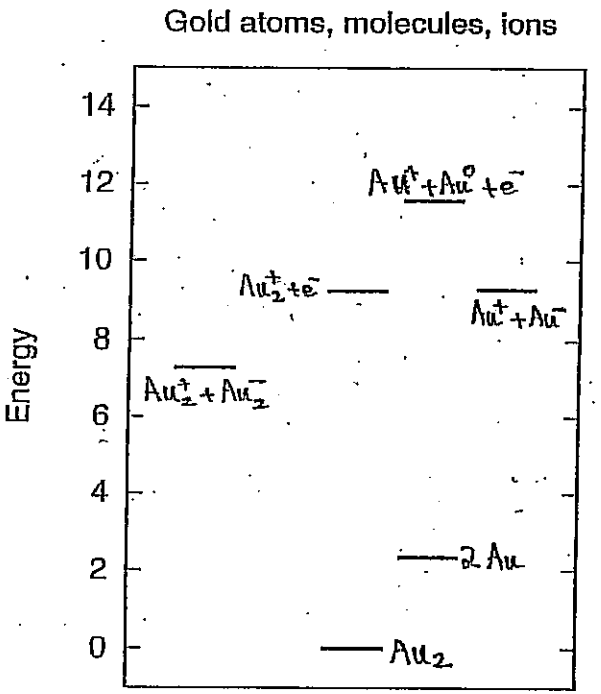
Conduction by charge transfer (n-type, p-type)

Semiconductor-metal transition? Sheath layers?

Photodetachment/photoconductivity

Radiation source?

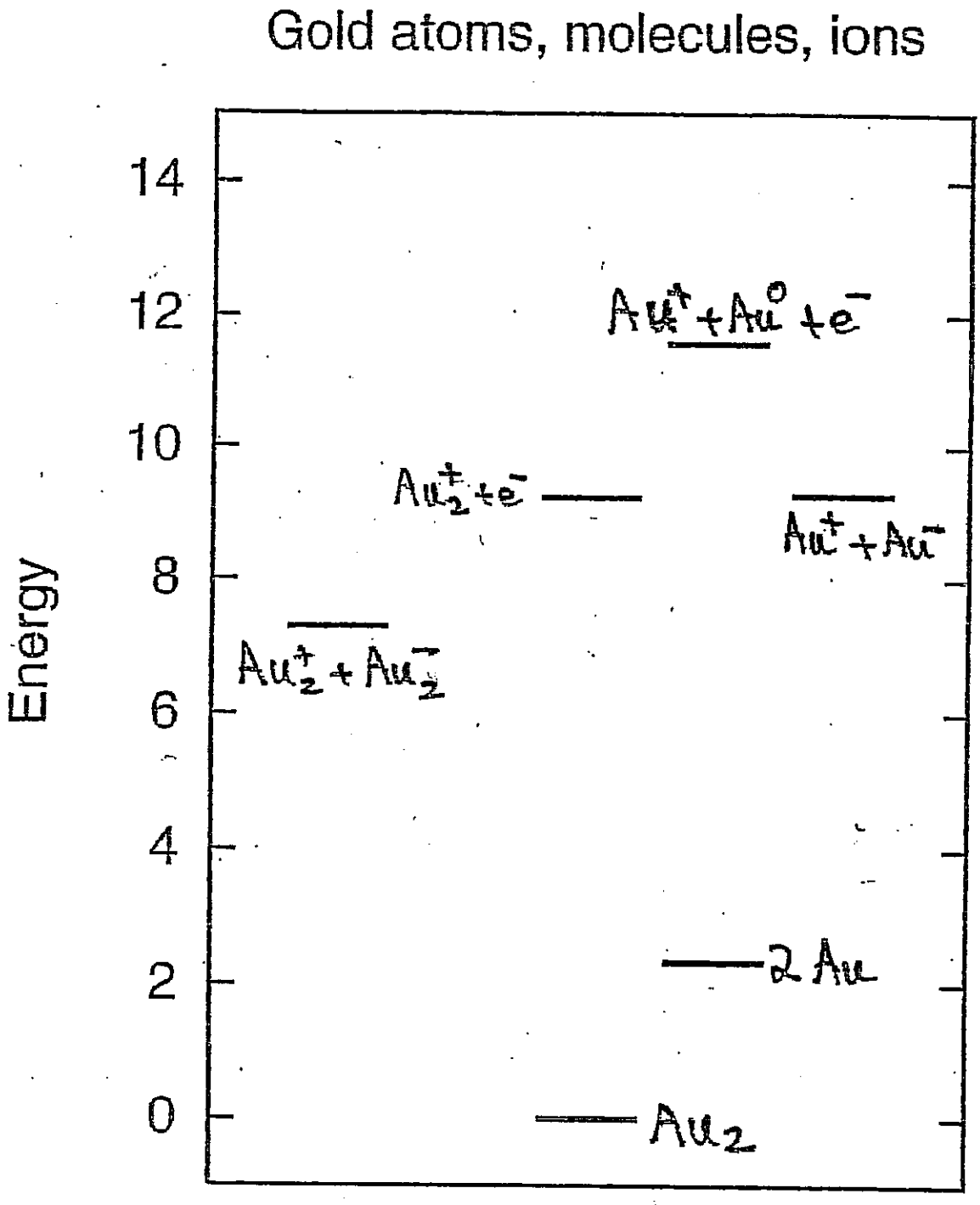
Nonequilibrium heating favors the ± plasma

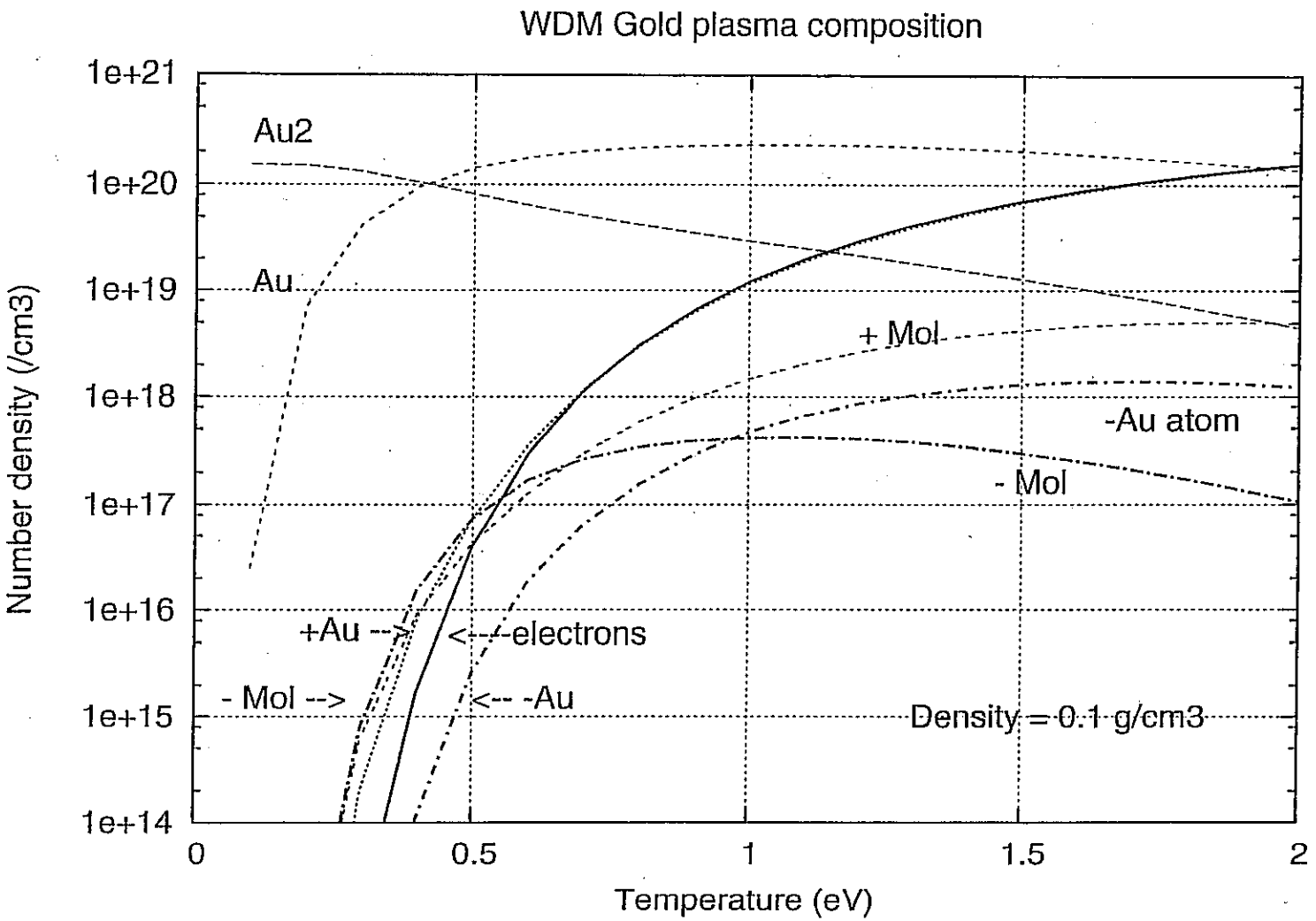



***Metals*** with strong electron affinity (Au, Sn) seem to show this. (Yoneda et al.)

***Covalent halogens*** ($Br_2$, $I_2$) are good candidates (L. Grisham & LBNL group)

PRL
JQSRT


Gold atoms, molecules, ions
Energy
14
12
10
8
6
4
2
0
$Au^+ + Au^0 + e^-$
$Au_2^+ + e^-$
$Au^+ + Au^-$
$Au_2^+ + Au_2^-$
2 Au
$Au_2$


WDM Gold plasma composition
Number density (/cm3)
1e+21
1e+20
1e+19
1e+18
1e+17
1e+16
1e+15
1e+14
0
0.5
1
1.5
2
Temperature (eV)
Au2
Au
+ Mol
-Au atom
- Mol
+Au -->
<-- -electrons
- Mol -->
<-- -Au
Density = 0.1 g/cm3

# *Metal-Insulator transition*

[ low σ --> high σ ]

SSP: Anderson, Mott, Thouless Datta Libby
Hensel, Faber, Endo, Kitamura
Hg, Cs (low Tc metals) have been studied in detail

WDM: De Silva, Benage, Yoneda - experiments
Laughlin, More, Busquet, Desjarlais (QMD)

At high densities, gap-closing or a percolation threshhold.

M. Desjarlais, Cont. Plasma Phys 41 (2001)

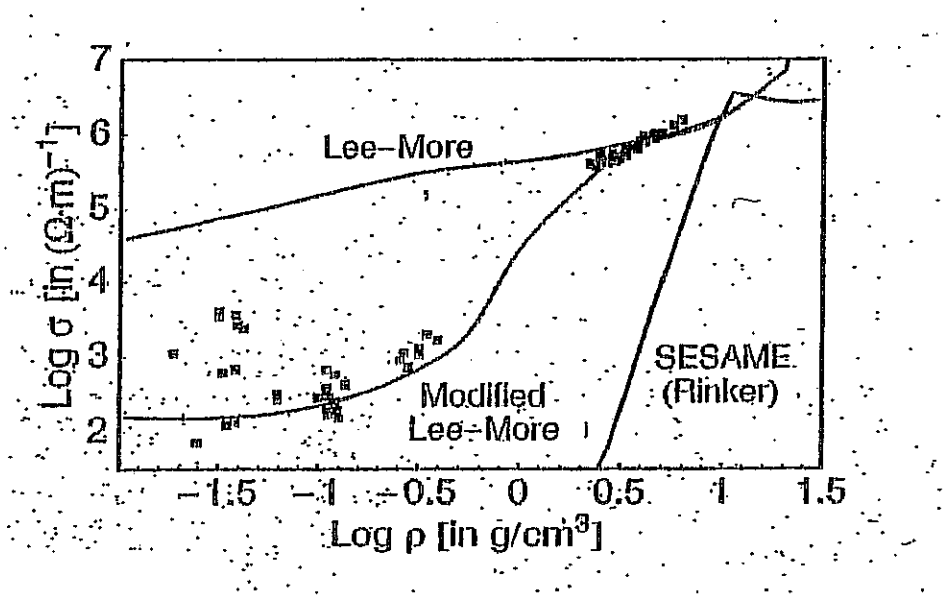
Big questions:

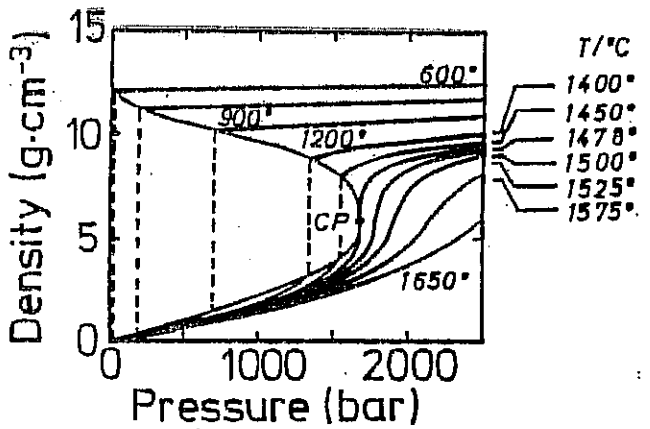
*Is there a sharp transition?*

*Where are the ρ,T boundaries?*

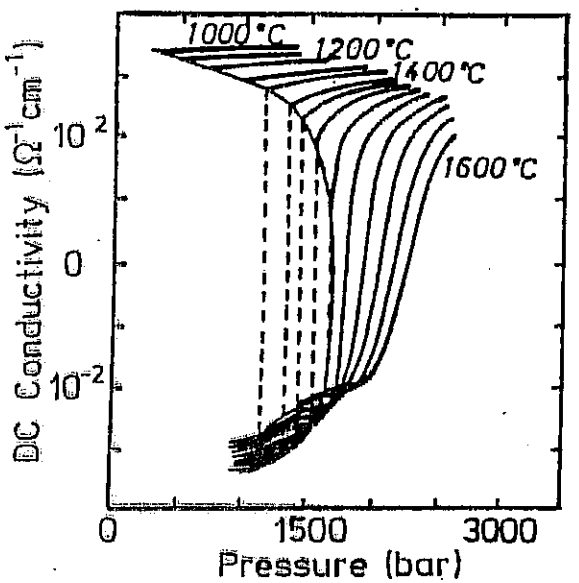
*What is the mechanism?*

polarons? Mott? Anderson?






(a)




(b)

Fig. 2.4. Isothermal equation of state (*a*) and DC electrical conductivity (*b*) data for mercury at sub- and supercritical temperatures as functions of pressure.



## 2.2 Fluid Semiconductors

### 2.2.1 *Covalent Bonding and Molecular Structure*

Fluid metals are not the only fluids in which the thermodynamic state strongly influences the electronic structure. When covalent bonding produces extended network or polymeric structures in the liquid state, the interparticle forces can depend sensitively on the density and temperature. Strictly speaking, the electronic structure of any liquid polymer is state-dependent since the vapor phase consists of monomers or other small species. This state-dependence takes on unusual characteristics, however, if there is significant electronic conductivity. In such a case there is again an electronic transition that accompanies the liquid-vapor transition.

The best elemental example is selenium which melts to form a liquid composed of extended chain molecules (Misawa and Suzuki, 1977). Twofold atomic coordination in these chains is associated with the bonding scheme shown in Fig. 2.5 (see, e.g., Cutler, 1977). The highest occupied electronic states forming a nonbonding or "lone-pair" band are separated from the unoccupied anti-bonding states by an energy gap $E_g$. It turns out that for selenium, $E_g$ is low enough that a significant number of electrons can be thermally excited to empty states in the upper band or to "acceptor" states associated with "defects" such as broken

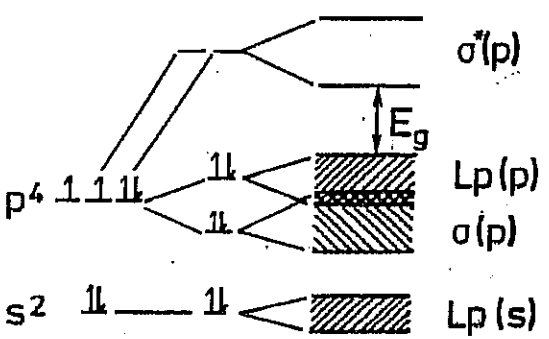



Fig. 2.5. Schematic electronic energy level diagram for selenium showing, from left to right, valence electron states of the free atom, splittings due to interactions with a second atom, and energy bands in the condensed phases of selenium. The $p$-electron bonding and antibonding states are denoted $\sigma(p)$ and $\sigma^*(p)$, respectively; Lp($s$) and Lp($p$) denote the lone-pair states for $s$- and $p$-electrons, respectively.

## *"Black glass"*

$\mathrm{Im}(\varepsilon) > \mathrm{Re}(\varepsilon) > 0$

$SiO_2$ $Al_2O_3$ $MgF_2$ have Ne-like closed-shell configurations.

Large energy gap (2s-2p to 3s-3p) == > transparent insulators.
At WDM temperatures, electrons can be excited 2p --> 3s
-- > long-lived holes in 2p states

Holes permit intra-atomic absorption 2s -- > 2p

***Experimental indications of this phenomenon:***

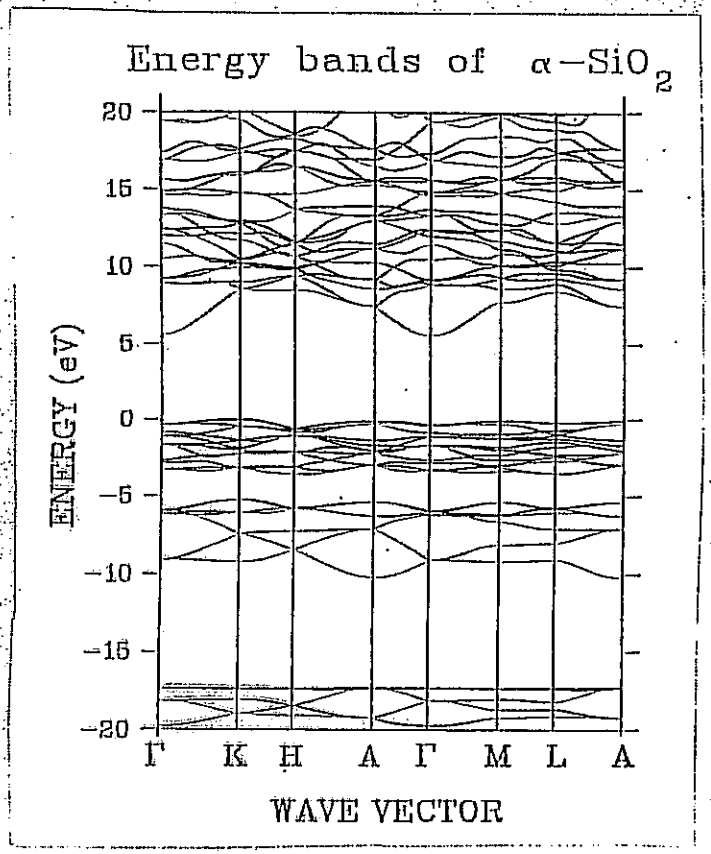
D. Price, Livermore -- USP laser absorption '95

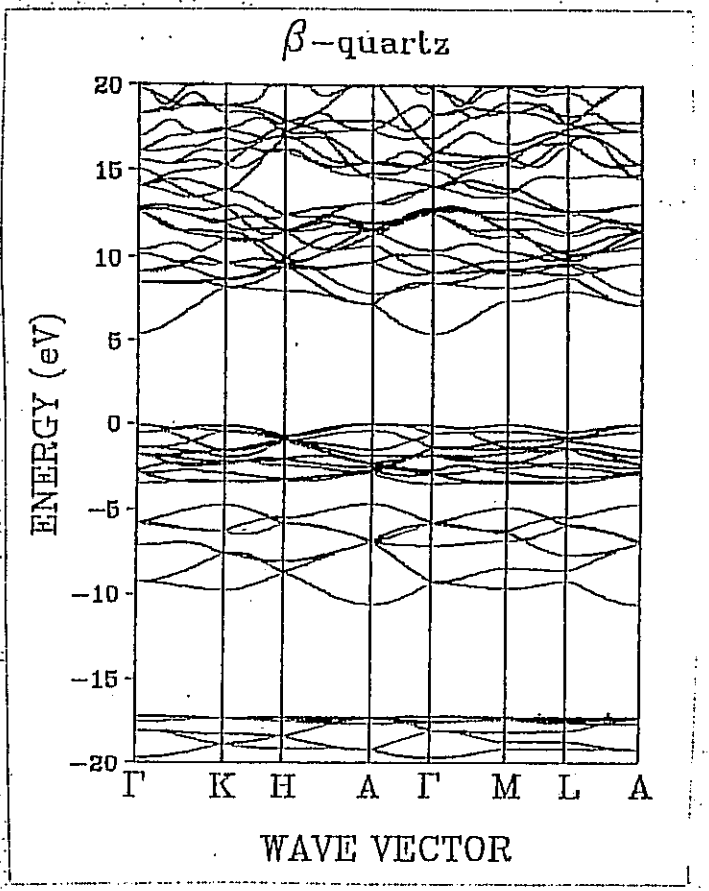
T. Tanabe, Kyushu University -- reactor experiment '03

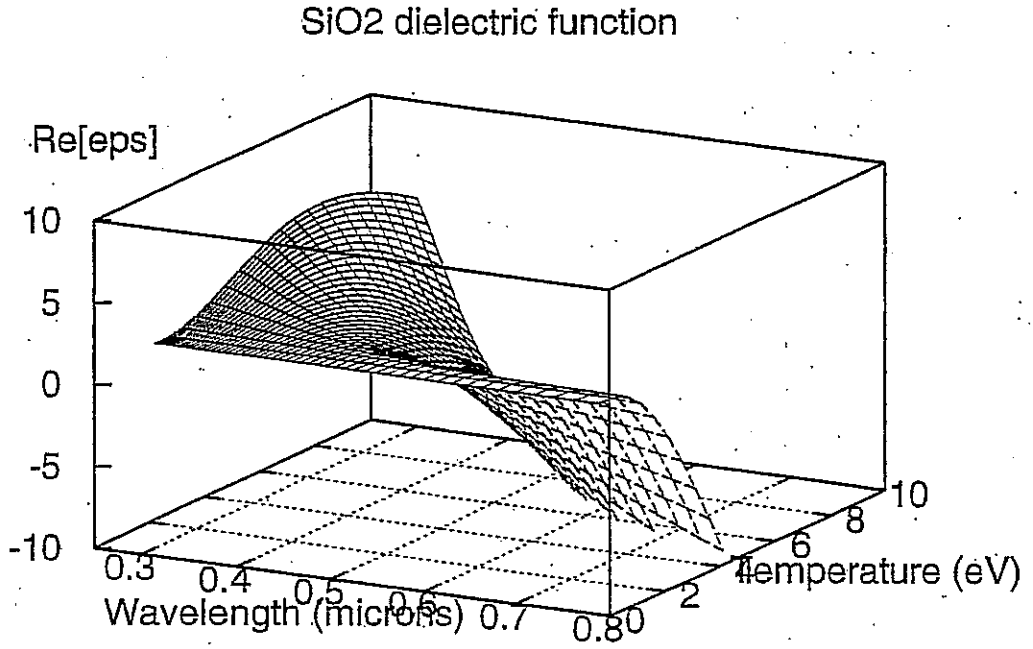
P. Renaudin, CEA Bruyeres -- sapphire window emission '04

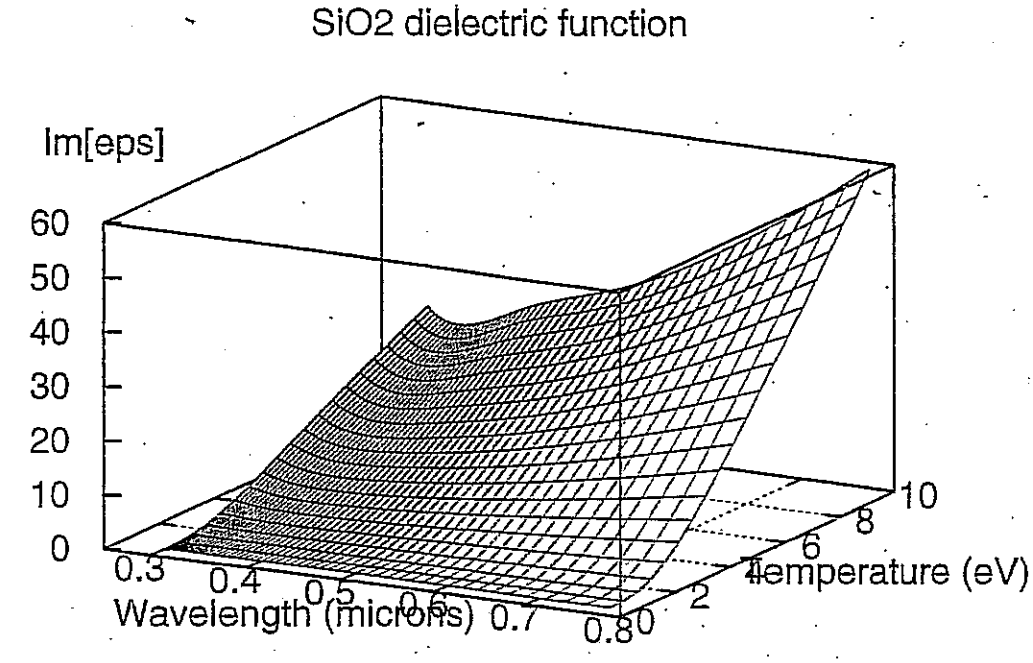
P. Lyons, F. Bieniosek - transient darkening of fibers '85

H. Yoneda - Laser and accelerator experiments Dec '06


Energy bands of α−SiO2
ENERGY (eV)
20
15
10
5
0
−5
−10
−15
−20
Γ K H A Γ M L A
WAVE VECTOR


β−quartz
ENERGY (eV)
20
15
10
5
0
−5
−10
−15
−20
Γ K H A Γ M L A
WAVE VECTOR


SiO2 dielectric function
Re[eps]
10
5
0
-5
-10
0.3
0.4
0.5
0.6
0.7
0.8
Wavelength (microns)
2
4
6
8
10
Temperature (eV)

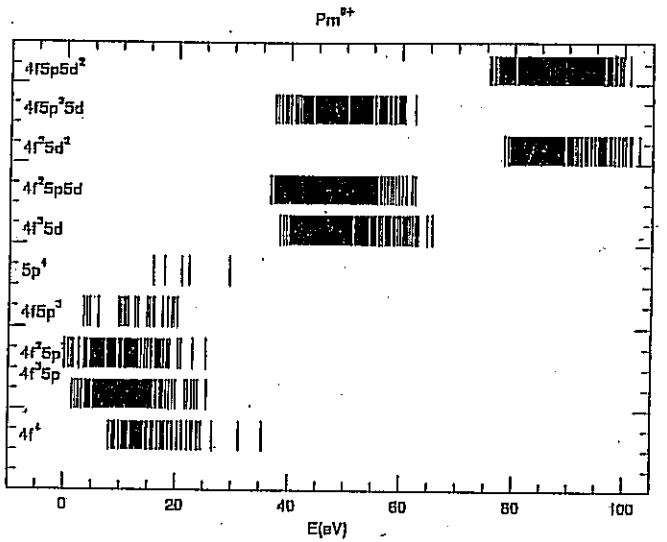

SiO2 dielectric function
Im[eps]
60
50
40
30
20
10
0
0.3
0.4
0.5
0.6
0.7
0.8
Wavelength (microns)
2
4
6
8
10
Temperature (eV)

## ***Shell-crossing and mixed valence*** [e.g., $Pm^{9+}$]

Theory predicts one-electron level crossing in rare earth ions.

Example:

5p-4f crossing in $Pm^{9+}$
I. Murakami, NIFS
HULLAC code




***Consequences:***

Large-scale ("Massive") configuration interaction { $4f^{m-n}\ 5p^{n}$ }

Anomalous broadband "white-light" emission
- *observed by Dublin laser spectroscopy group*

This could set the upper limit of WDM

# *MODELING TOOLS TO STUDY WDM*

R. More, T. Kato, H. Yoneda

J. Wurtele, J. Barnard, G. Penn, A. Friedman

- o New **EOS codes**
- o Planar **hydrodynamic code** "DPC"
- o **Electromagnetic wave-solver** for pump-probe laser ellipsometry
- o **Material models** for Au, Sn, W, glass, $Br_2$, etc.
- o Monte Carlo simulation of metal-insulator transition
- o **Foil heating simulations** using LLNL Hydra code. Foam targets.
- o **Beam-target interaction** codes for ion beam propagation and heating

# Warm dense matter and the liquid-vapor transition

**1st generation:** Hydrodynamics with equilibrium EOS

Two-phase EOS with Maxwell construction

**2nd generation:** Evaporation kinetics, surface tension, yield strength

Evaporation / condensation

fast at high ρ, high T

slower at low ρ, low T

"Spinodal" region has a micro-instability when $\frac{\partial p}{\partial \rho} < 0$

***For hot metals, EVERYTHING should be tested by experiment.***

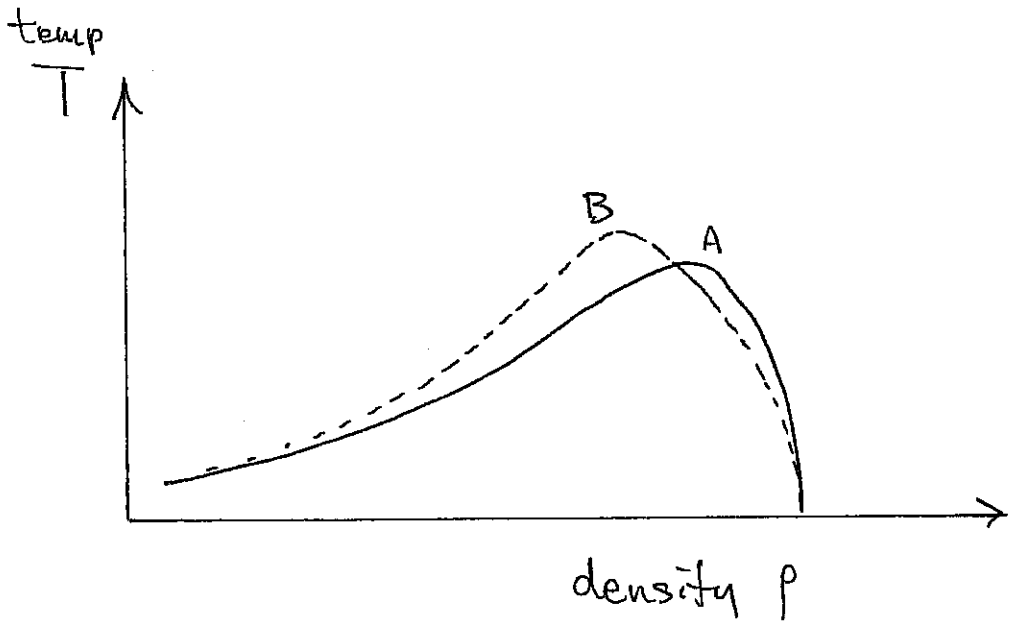
QUESTION:

***Do excitation and ionization distort the 2-phase boundary ?***

Tungsten is a candidate:

$T_c$ = 1.1 to 1.5 eV

IP = 7.98 eV

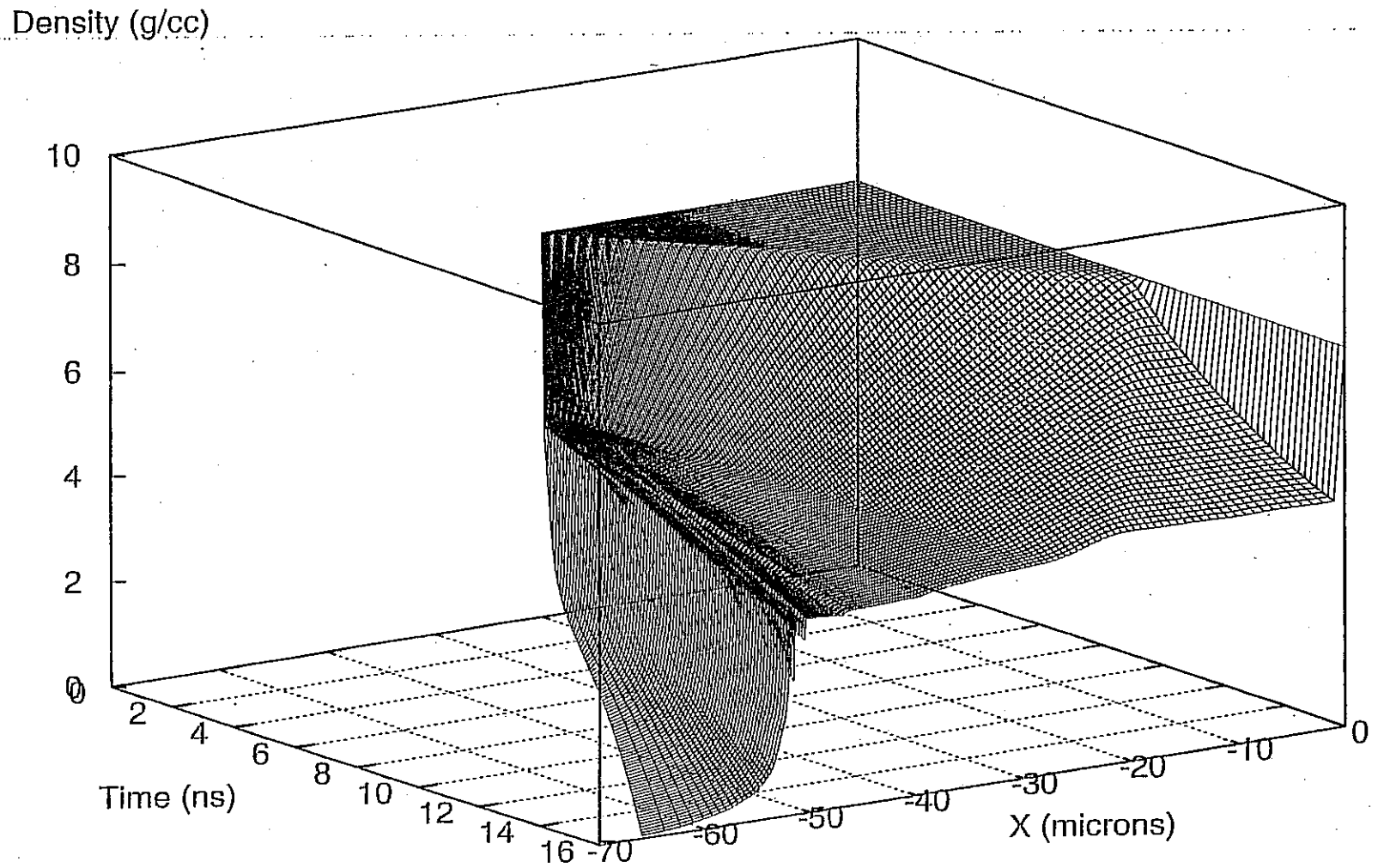



New Tungsten EOS constrained by QMD data from M. P. Desjarlais (SNLA)

Atom pair-potential V(R) changes with excitation, ionization.

For these materials, the fluid "corresponding states" EOS is invalid.

Sn release from solid-density (To = 1 eV)



# *Hydrodynamic release into liquid-vapor region:*

accurate EOS + good resolution + robust EOS look-up

Release from a uniform initial state: More, Kato, Yoneda find ***shelf structure***.

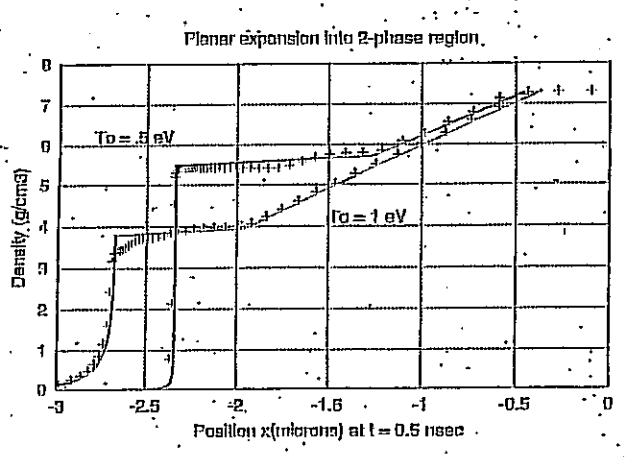
- o Sharp interface of liquid-metal region
- o Low-density (2-Φ) precursor

Numerical simulation *agrees with* exact analytic solution

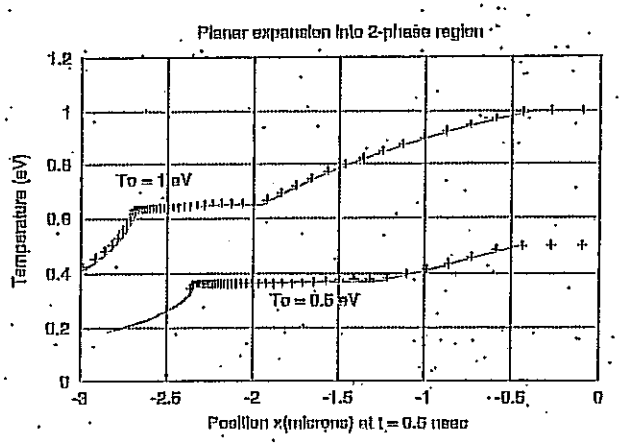
(*) also agrees with Nishihara particle simulation

----- = Exact solution  -+-+-+- = Numerical hydrodynamics

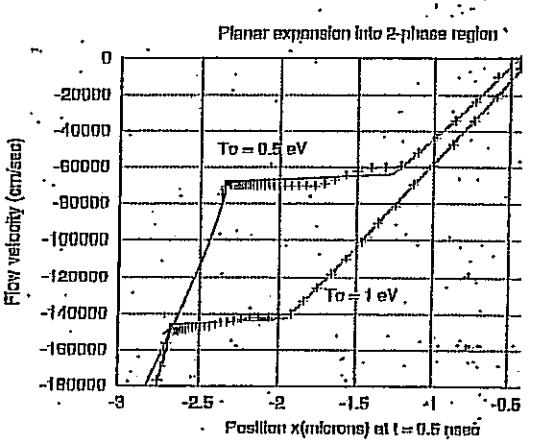
DENSITY




TEMPERATURE




FLOW SPEED

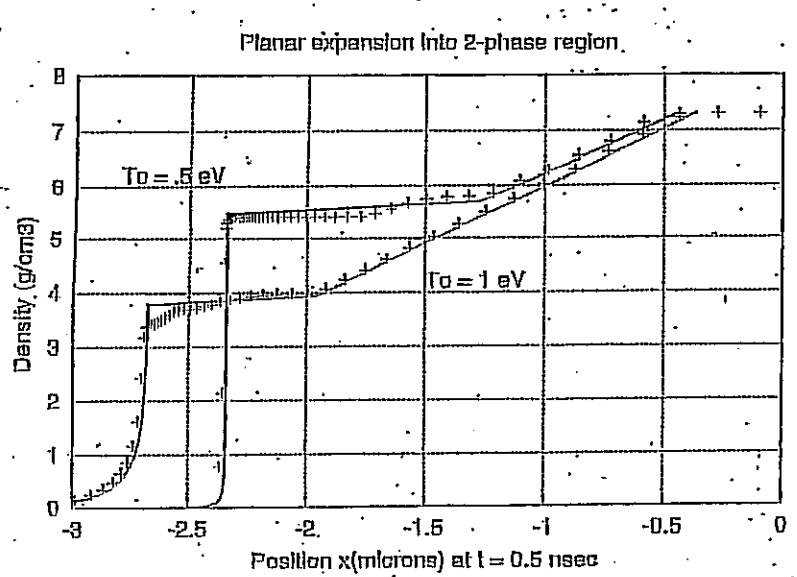


# Numerical simulation agrees with exact analytic solution:

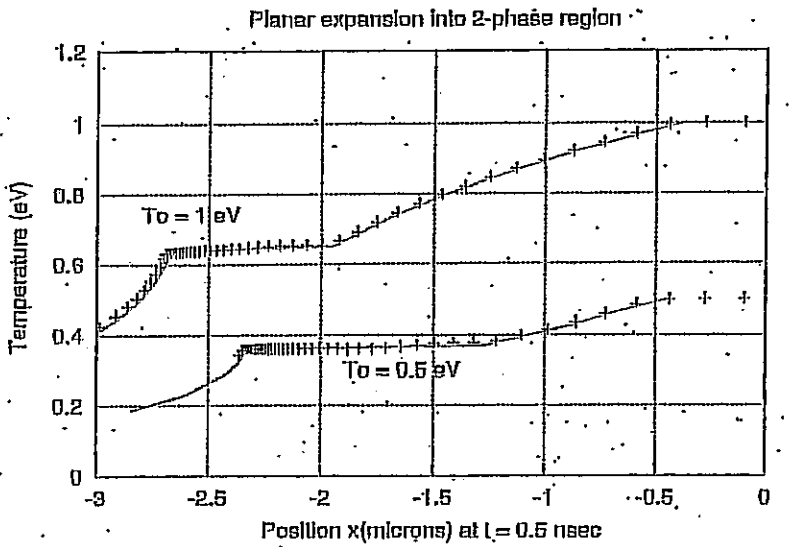
----- = Exact solution

-+-+-+- = Numerical hydrodynamics

## DENSITY




## TEMPERATURE



## *Many applications of WDM information:*

Fusion research -

- Pulsed power - electrical heating and wire dynamics
- ICF - early-time behavior; scale-up pathway for HIF; debris
- MFE - Pellet injection dynamics; wall interactions

Radiation sources - EUV

Laser cutting and machining

Plasma processing, high-temperature chemistry

Future high-power semiconductors?

Geophysics, astrophysics, condensed-matter

# MAKING WARM DENSE MATTER:

## *Electric current heating* *(since 1920)*

| | |
|---|---|
| I(t), V(t) -- > know energy input | rise time > 10 - 50 nsec |
| Energy in WDM range | expansion -- > nonuniform |
| Inexpensive sources | conductivity, deposition change |

## *Laser heating* *(since 1970)*

| | |
|---|---|
| Wide range of temperatures | Deposition at low density |
| Short pulse -- > high density | Absorption changes with T |
| High rep-rate -- > data rich | Needs hydro modeling |
| Fast diagnostics (pump-probe) | nonequilibrium |
| Large and small facilities exist today | competition for laser time |

## *Accelerator heating* *(today)*

| | |
|---|---|
| Uniform deposition over ~ 1 micron | Power limit in today's machines |
| Robust deposition, any target | Need space-time focusing |
| Foam targets with uniform deposition | Need diagnostics |

## *X-ray heating* *(future)*

***Real discoveries will be verified several ways***

# *In WDM there are new things to learn*

*New phenomena*

*limited range of T  ==>  temperature flags*

*Technologies for WDM research:*

*Heating - X-rays, shock release, lasers, electrical, e-beam,* ***and accelerators***

*Diagnostics - hydro speed, electrical, optical, UV and X-ray transmission*

*Modeling, theory - Hydro codes, EOS, conductivity, atomic kinetics, QMD*

## *WDM research is "in construction"*

Basic phenomena  Basic organization