WARM DENSE MATTER RESEARCH

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J. Wurtele, G. Penn, G. Logan (LBNL), J. Barnard, A. Friedman (LLNL)

- **o Remarkable phenomena in WDM** < -- *What's there in WDM?*
- **o Tools and applications** < -- *Sketch of today's research*
- **o 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 ==> rapid hydro ==> dynamic experiments

"*Warm*" -- >

Melt and boil anything, even W ==> 2-phase (liquid-vapor)

Electronic excitation and maybe ionization

but not high charge, not much radiation, not CR

"*Dense*" -- >

Atoms interact with an environment (disordered fluid environment)

"*Matter*" -- >

study material phases and properties, look for the patterns

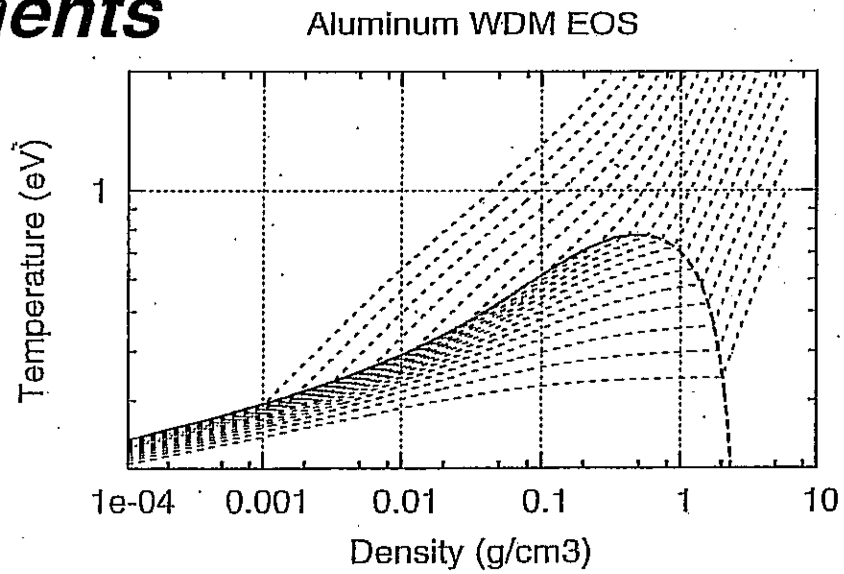
"***Research***" **implies we want to find and understand *new things*.**

Typical WDM experiments

A Heat slowly

B Heat rapidly

C Shock heat & compress



Time before disassembly $\tau \sim L / c_s \sim (1\ \mu) / (10^5$ 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

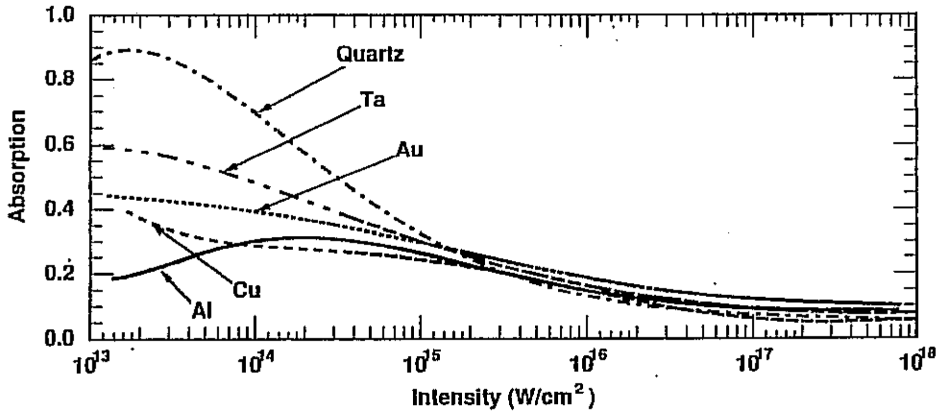
Aluminum is well-described by "usual" plasma theory (QEOS + Drude)

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.

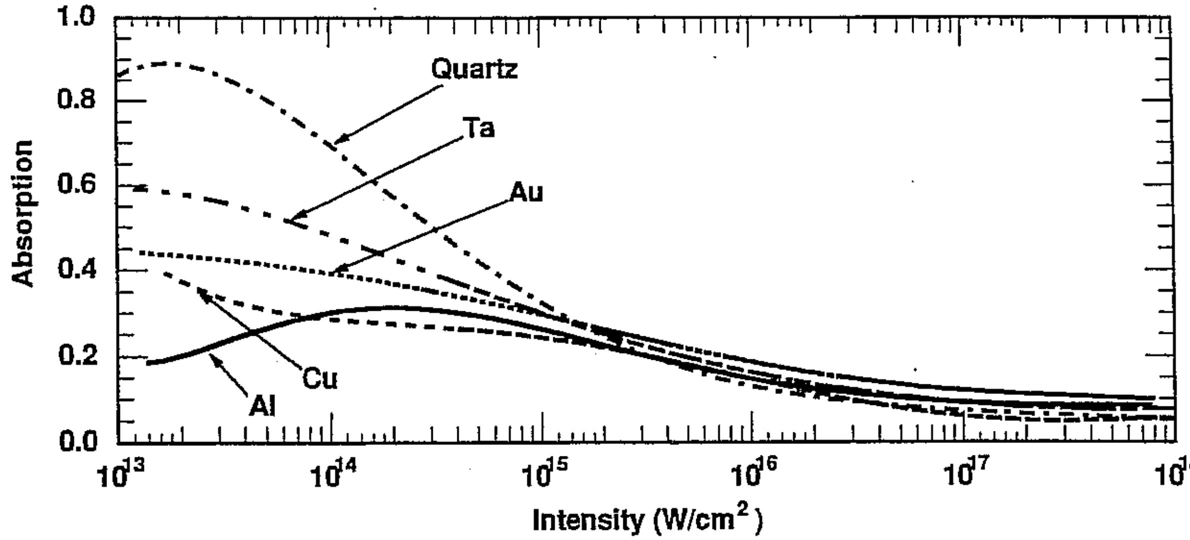


FIG. 1. Absorption fraction vs peak laser intensity for aluminum, copper, gold, tantalum, and quartz targets. In Figs. 1, 3, 4, and 5 laser intensity is the temporal and spatial peak value of the laser intensity.

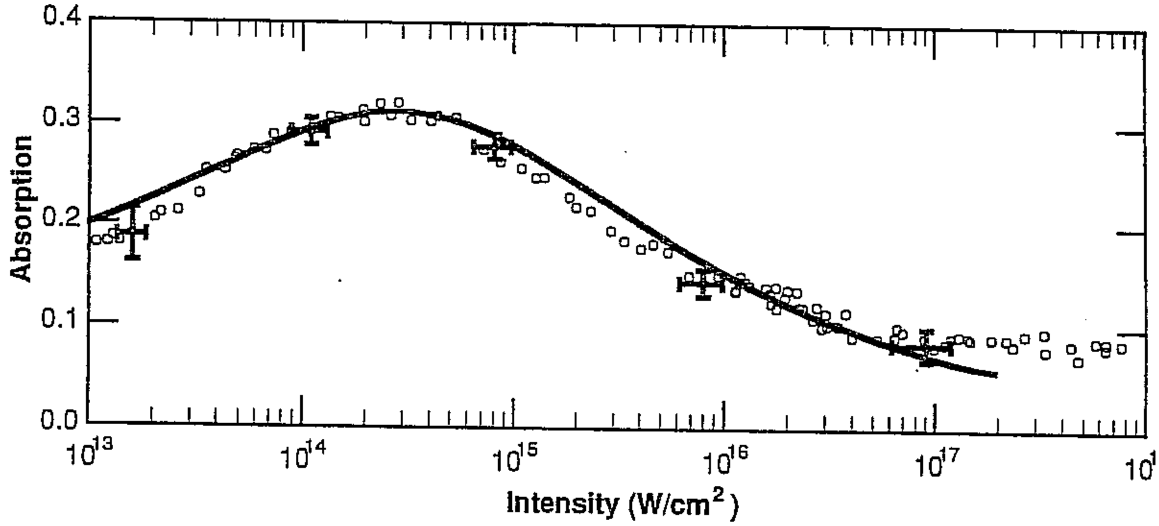


FIG. 5. Measured and calculated absorption fractions for an aluminum target vs peak laser intensity. The calculation assumes a Gaussian laser spatial profile. Error bars shown indicate systematic uncertainty in intensity and random error in absorption. The absolute absorption scale is believed to be known to ± 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_ν, 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_ν	$=$	$\varepsilon(\nu, \rho, T)\, B_\nu(T)$	$\varepsilon \leq 1$, $\varepsilon = a$
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 σ --> low σ]
- o "Black glass" [$\mathrm{Im}(\varepsilon)$ > $\mathrm{Re}(\varepsilon)$ > 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

Electronegative plasma predicted at WDM temperatures.

plus and minus ions   but n_e $\ll$ N_+

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

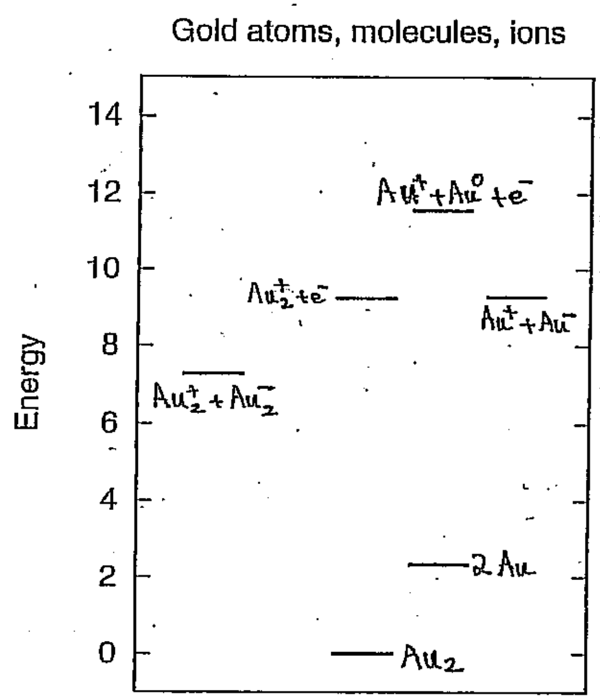
Conduction by charge exchange   (n-type, p-type)

Semiconductor-metal transition?   Sheath layers?

Photodetachment/photoconductivity

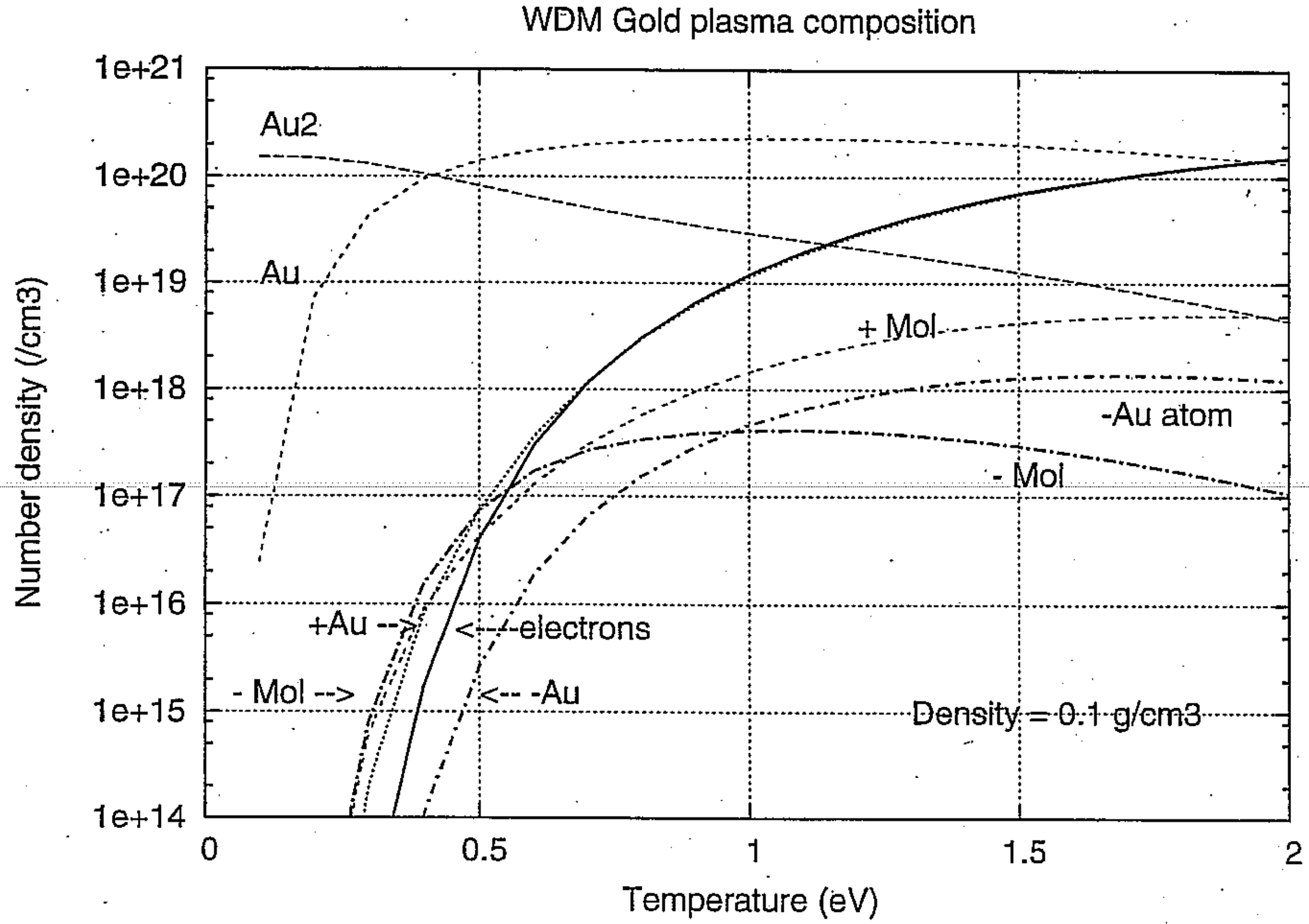
Radiation source?

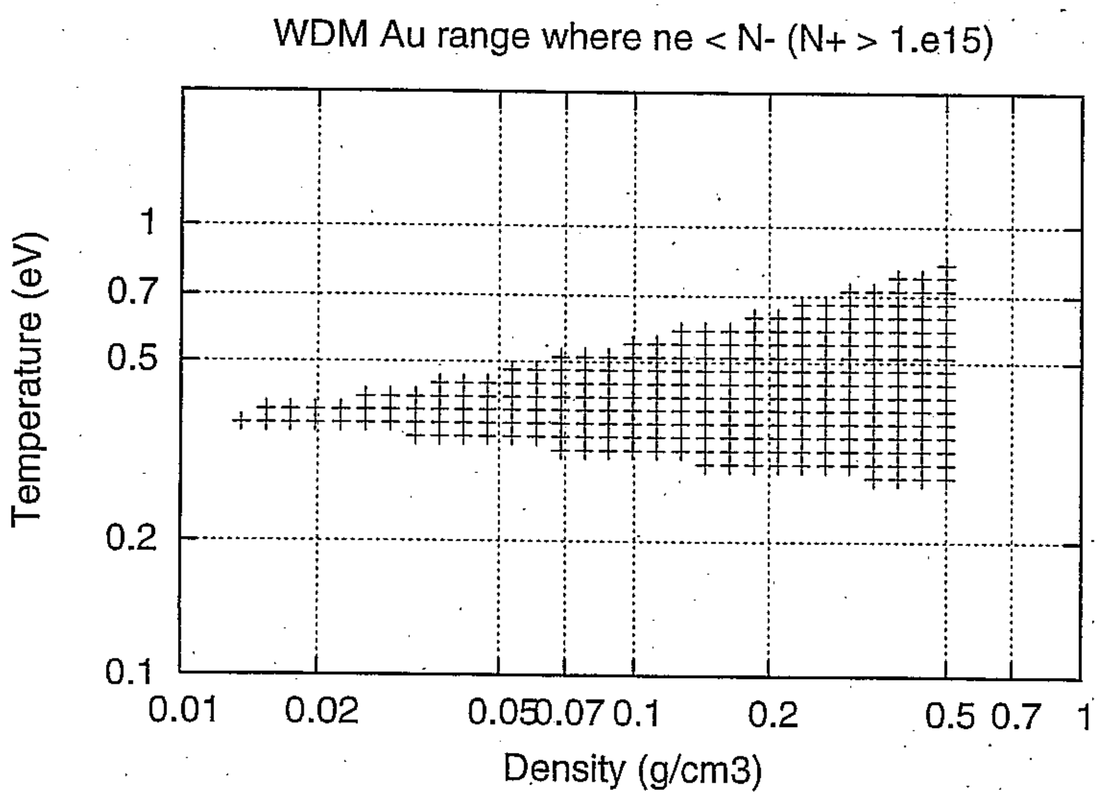
Nonequilibrium heating favors the ± plasma

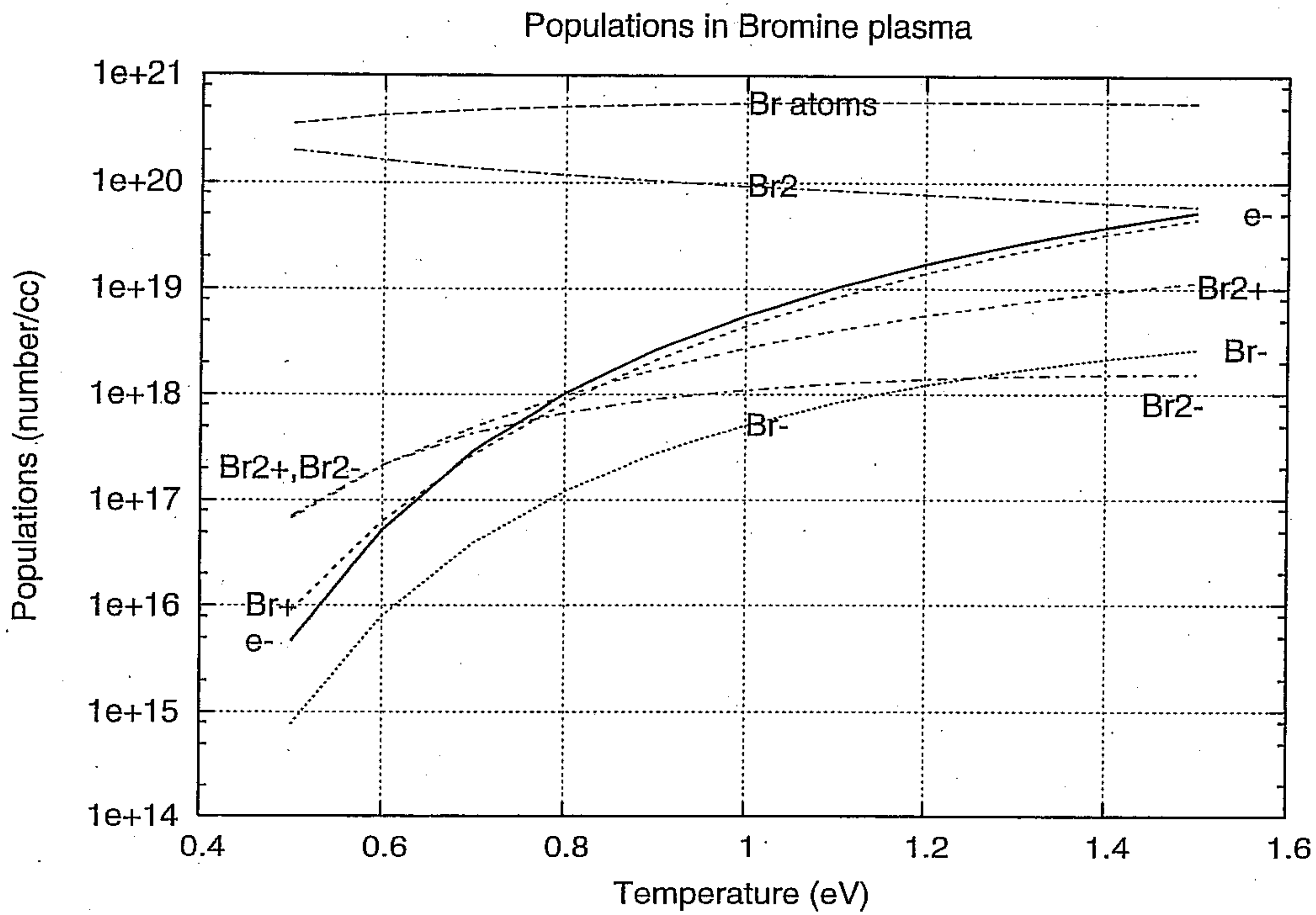


Metals with strong electron affinity (Au, Sn) seem to show this. (Yoneda et al.)

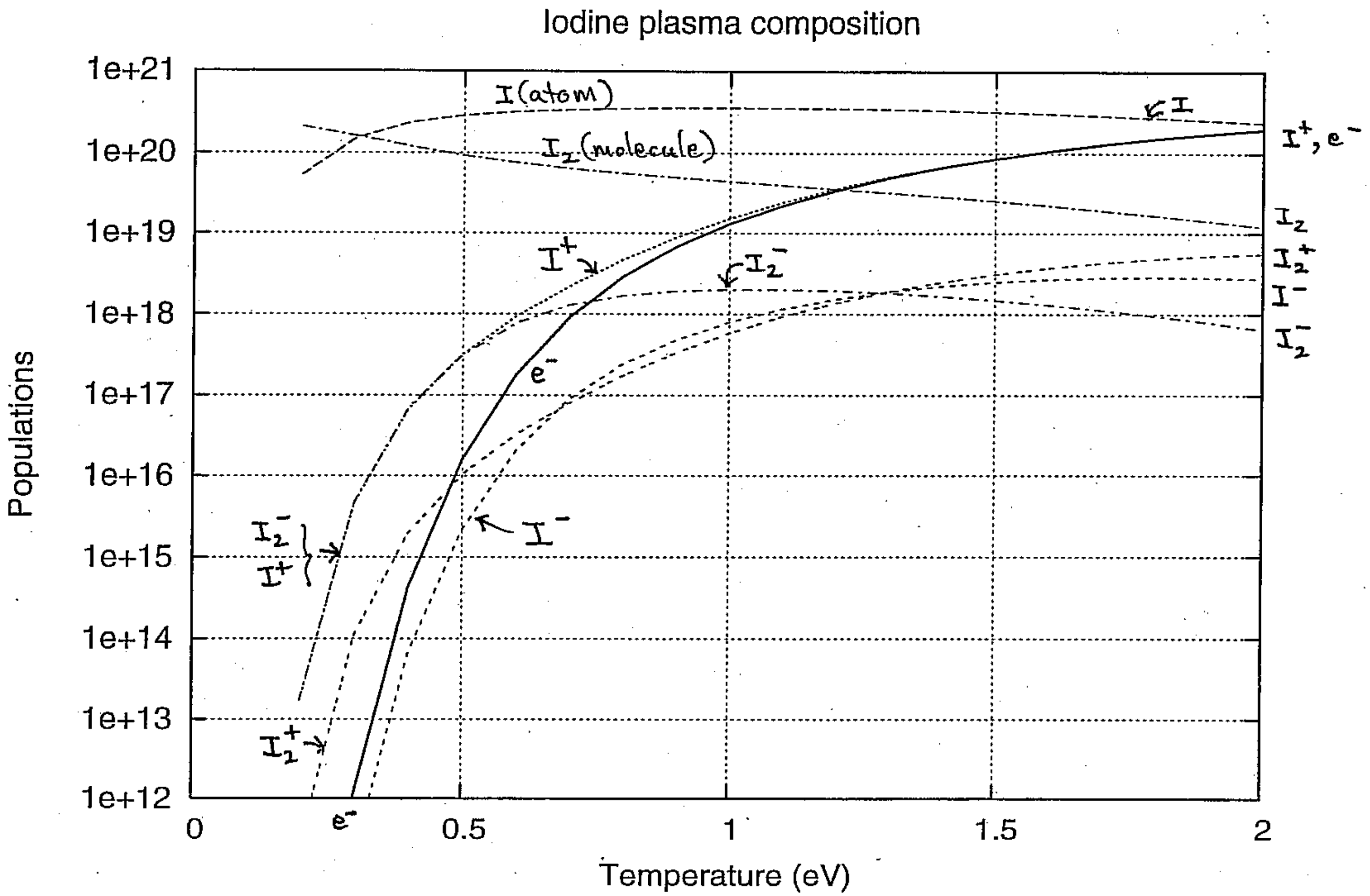
Covalent halogens (Br_2, I_2) are good candidates. (L. Grisham & VNL group)

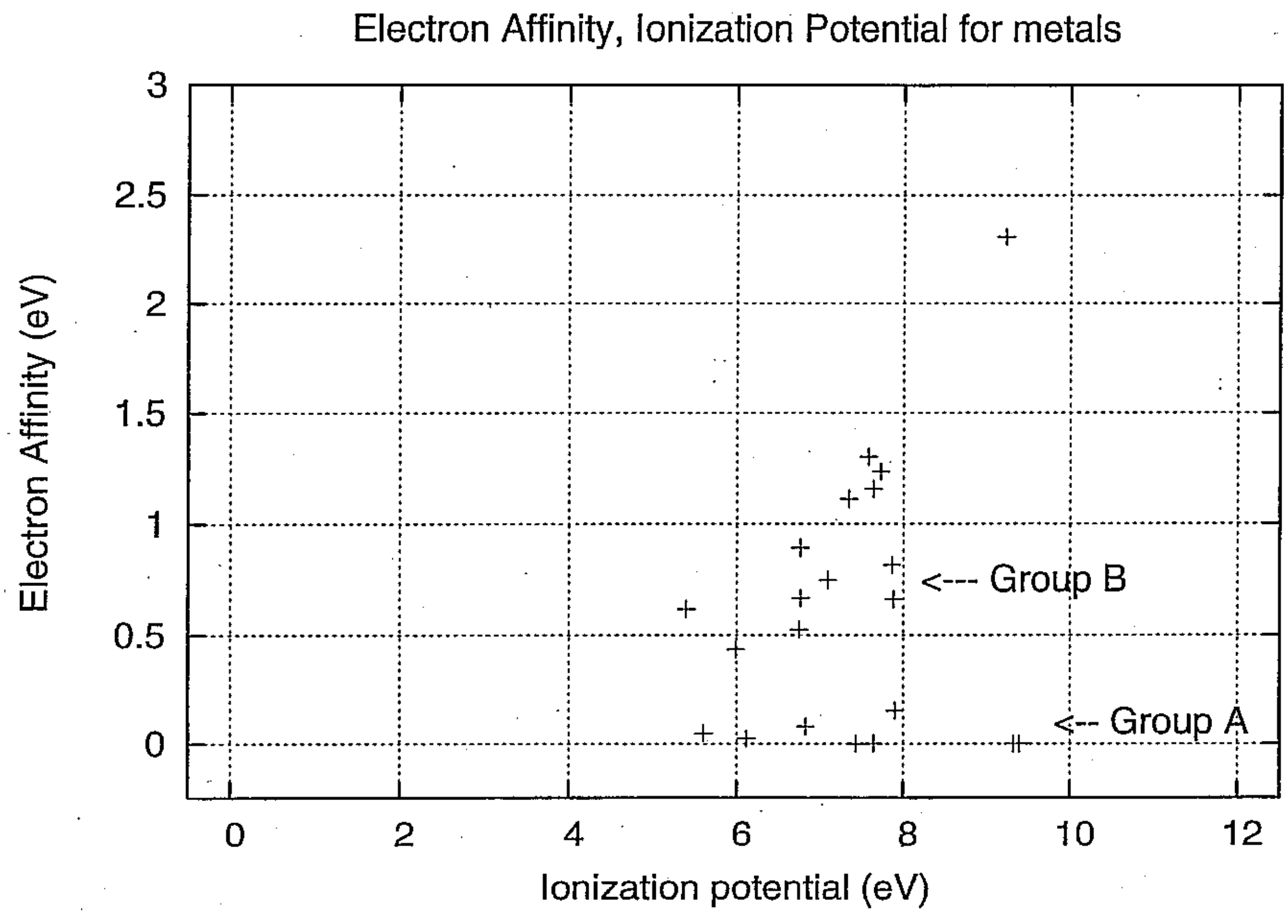

WDM Gold plasma composition
Number density (/cm3)
1e+21
1e+20
1e+19
1e+18
1e+17
1e+16
1e+15
1e+14
0
0.5
1
1.5
2
Temperature (eV)
Au2
Au
+ Mol
-Au atom
- Mol
+Au -->
<--- electrons
- Mol -->
<-- -Au
Density = 0.1 g/cm3


WDM Au range where ne < N- (N+ > 1.e15)
Temperature (eV)
1
0.7
0.5
0.2
0.1
0.01
0.02
0.05
0.07
0.1
0.2
0.5
0.7
1
Density (g/cm3)



Populations in Bromine plasma
Populations (number/cc)
Temperature (eV)
1e+21
1e+20
1e+19
1e+18
1e+17
1e+16
1e+15
1e+14
0.4
0.6
0.8
1
1.2
1.4
1.6
Br atoms
Br2
e-
Br2+
Br-
Br2-
Br-
Br2+,Br2-
Br+
e-


Iodine plasma composition
Populations
1e+21
1e+20
1e+19
1e+18
1e+17
1e+16
1e+15
1e+14
1e+13
1e+12
0
0.5
1
1.5
2
Temperature (eV)
I (atom)
I2 (molecule)
I
I+, e-
I2
I2+
I-
I2-
I+
e-
I2-
I+
I2+
e-



Electron Affinity, Ionization Potential for metals
3
2.5
2
1.5
1
0.5
0
Electron Affinity (eV)
0
2
4
6
8
10
12
Ionization potential (eV)
<--- Group B
<-- Group A

Metal-Insulator transition [low σ --> high σ]

SSP: Anderson, Mott, Thouless Datta
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:

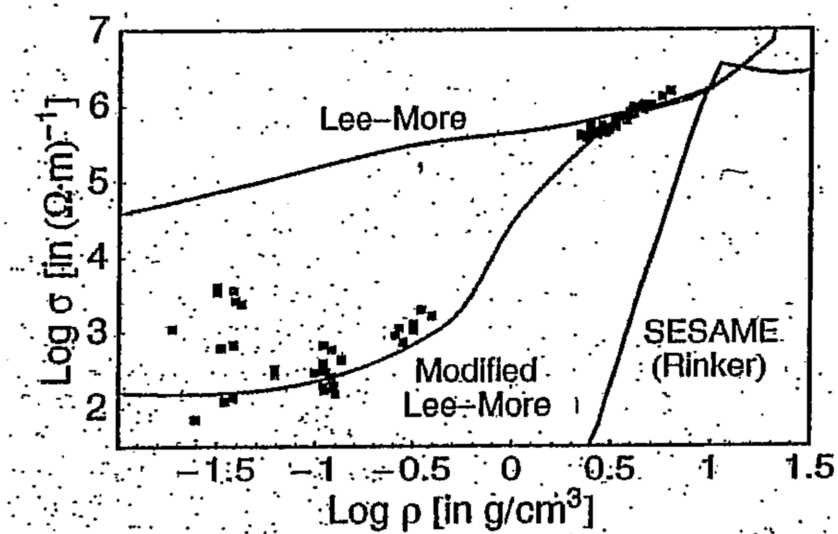
Is there a sharp transition?

Where are the ρ,T boundaries?

What is the mechanism?

polarons? Mott? Anderson?

Contrib. Plasma Phys. **41** (2001)

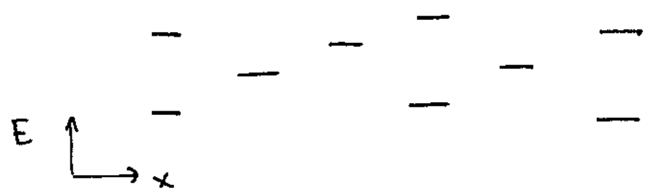


METAL-INSULATOR TRANSITION [low σ --> high σ]

Monte Carlo simulations for a line-width modulated MI transition

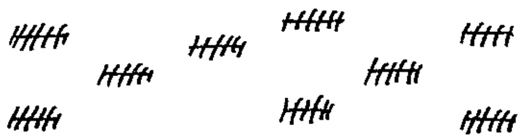
RMM, NIFS

low-σ state:



inhomogeneous disorder inhibits charge transfer

high-σ state:



homogeneous (collisional) widths enable charge transfer

Linewidth controls conduction

"Black glass"

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
leaving long-lived holes in 2p states (band minima, local potentials)

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

Experimental indications of this phenomenon:

D. Price, Livermore -- USP laser absorption

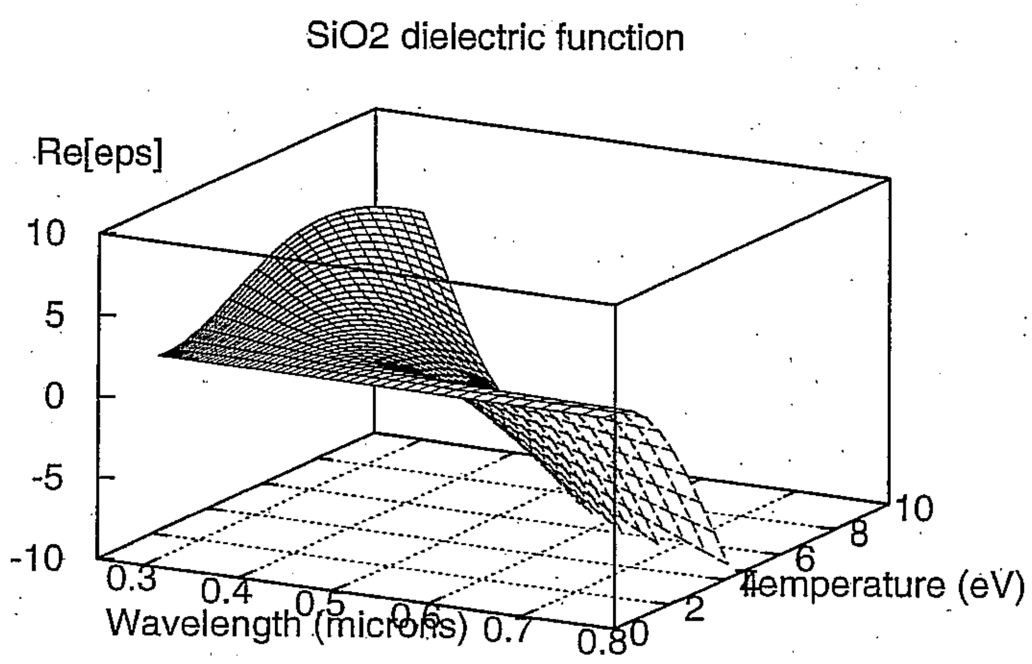
T. Tanabe, Kyushu University -- reactor experiment

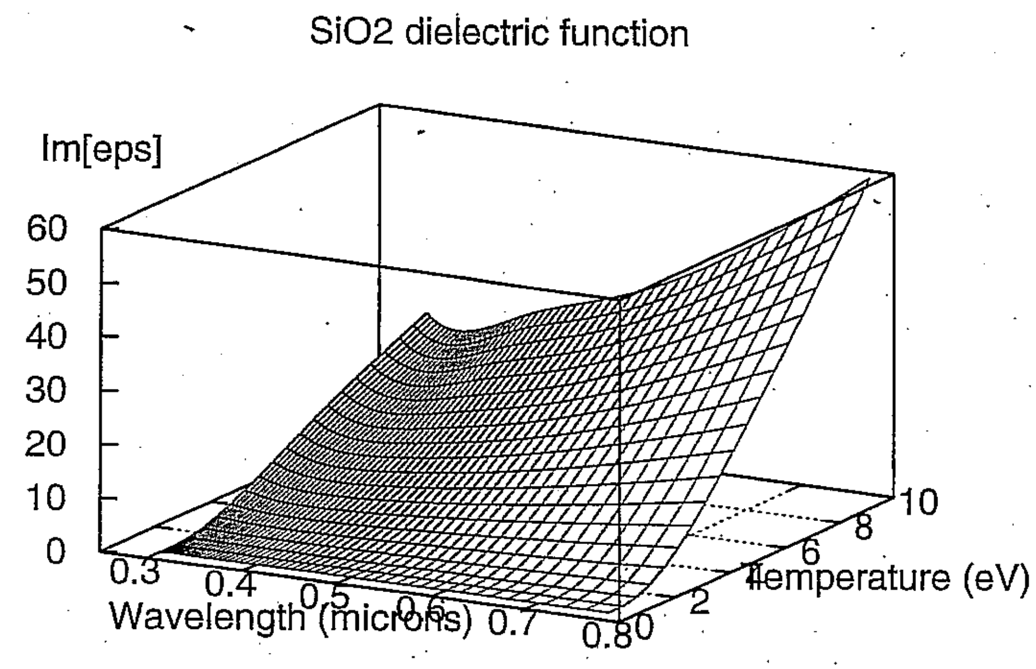
P. Renaudin, CEA Bruyeres -- sapphire emission spectrum

F. Bieniosek - LBL/LANL experiment

Signature: anomalous $\varepsilon(\omega)$ at WDM temperatures:

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


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

MODELING TOOLS TO STUDY WDM

R. More, T. Kato, H. Yoneda

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

o New **EOS code**

o Planar **hydrodynamic code**

o **Electromagnetic wave-solver** for laser pump-probe 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.   Collela code project

o **Beam-target interaction** codes for ion beam propagation and heating

Warm dense matter involves liquid-vapor transition

Two-phase EOS with Maxwell construction

Equilibrium is only the simplest possibility
Requires seeds for bubbles or droplets

Evaporation / condensation kinetics is fast at high ρ, high T

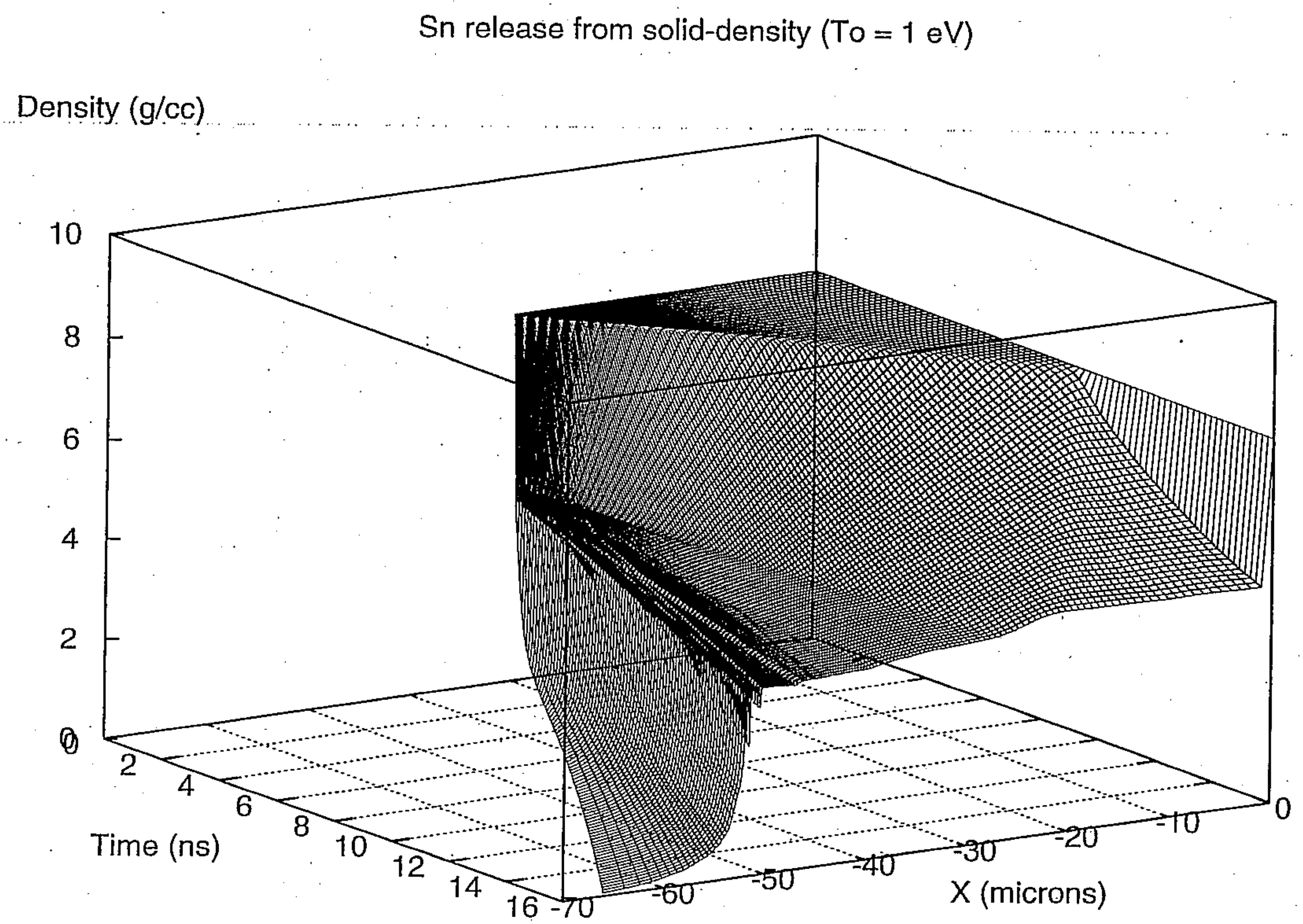
slower at low ρ, low T

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

1st generation: Hydrodynamics with equilibrium EOS (Maxwell)

2nd generation: Surface tension, yield strength, evaporation kinetics

EVERYTHING must be verified by experiment.


Sn release from solid-density (To = 1 eV)
Density (g/cc)
10
8
6
4
2
0
Time (ns)
2
4
6
8
10
12
14
16
X (microns)
-70
-60
-50
-40
-30
-20
-10
0

Hydrodynamic release into liquid-vapor region:

accurate EOS + good resolution + robust EOS look-up

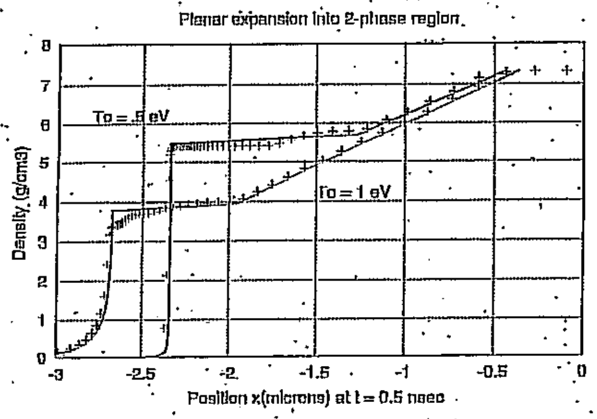
Release from a uniform initial state: More, Kato, Yoneda find ***shelf structure***.

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

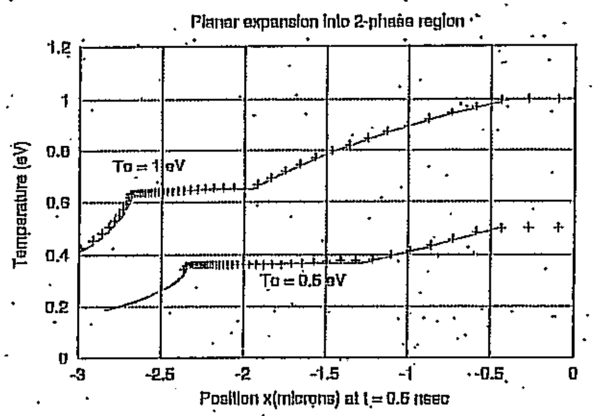
Numerical simulation *agrees with* exact analytic solution
(*) also agrees with Nishihara particle simulation

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

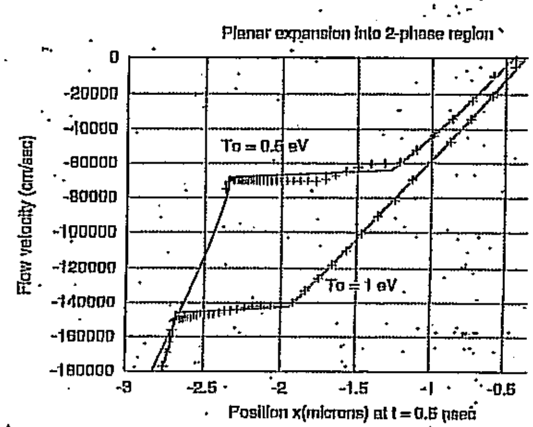
DENSITY



TEMPERATURE



FLOW SPEED



Shock-release experiments (M. Knudsen)

Two-flash signal when aluminum release hits LiF window.

Simulate Sn release from solid density 1 eV initial state

Low-density precursor	==>	first flash
Liquid-density layer	==>	second flash

Calculate with new aluminum EOS:

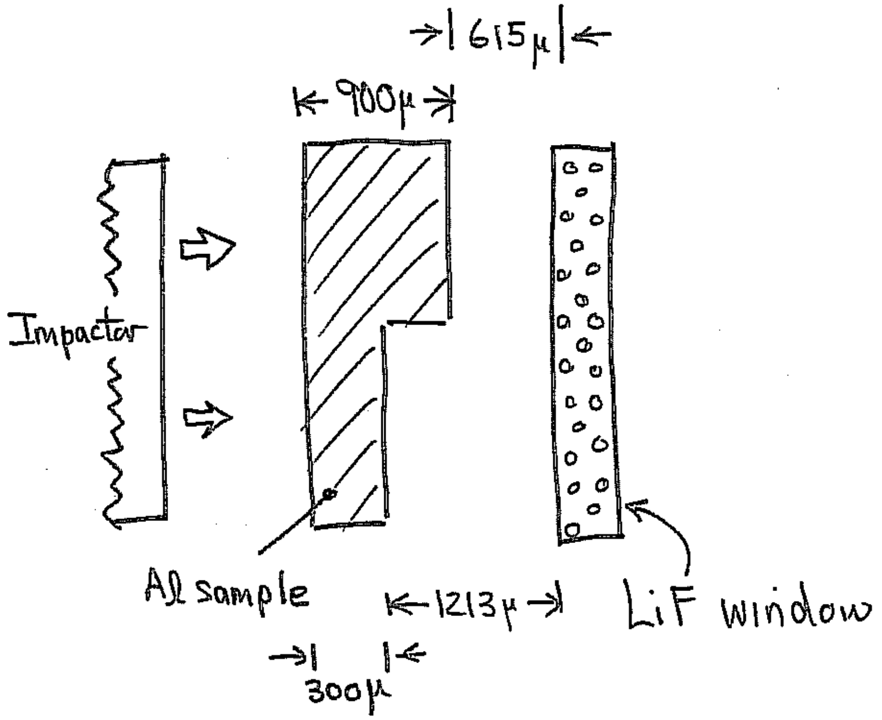
3 Speeds are *consistent* with SNLA experiment: u_s, v_{fast}, v_{slow}

Shock release, isochoric release both follow adiabats

Shock release material begins with particle velocity u_p

Window interaction blocks emission when liquid slug hits it.

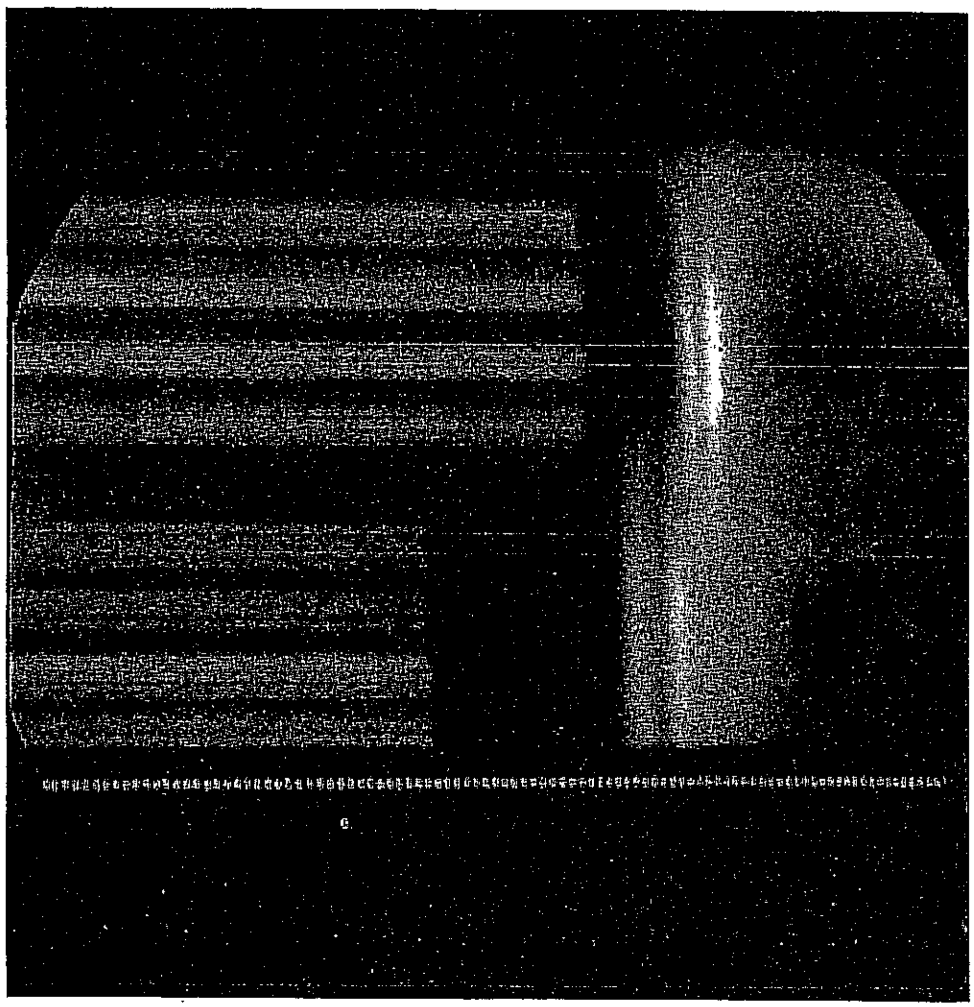
M. Knudsen, Shock-Release experiment

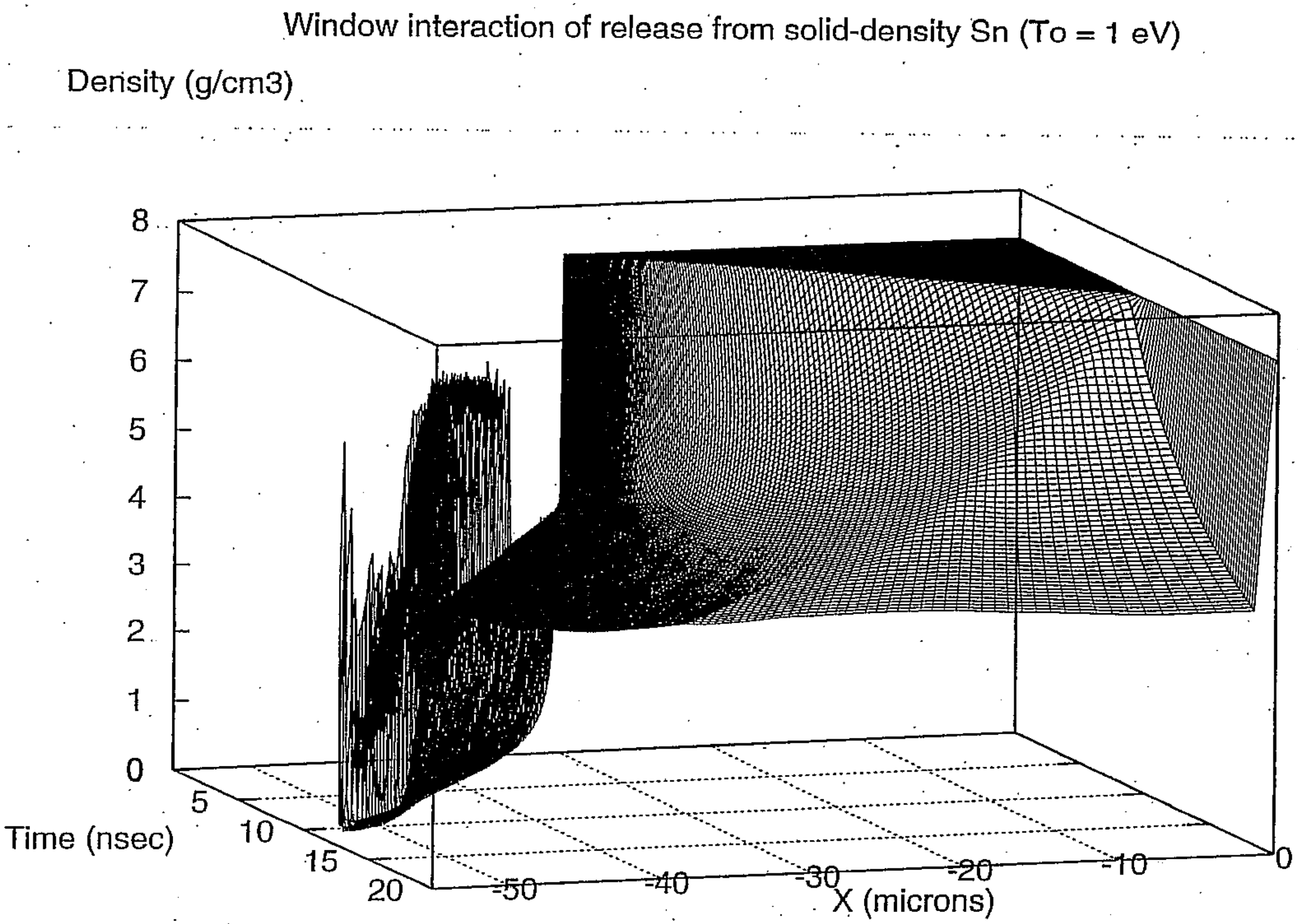


$u_s = 17.82 \frac{\mu}{nsec}$

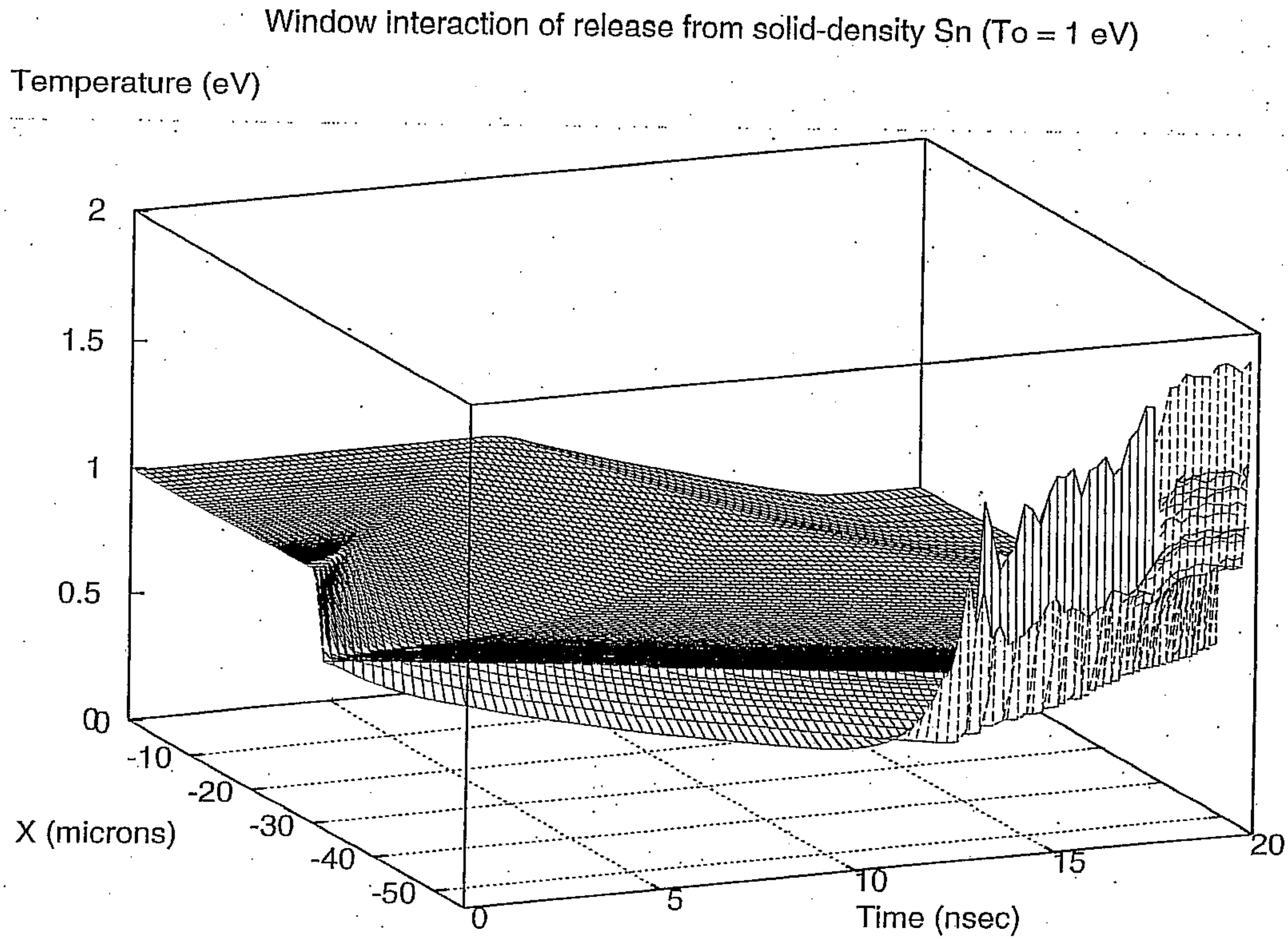
$u_p = 9.66 \frac{\mu}{nsec}$

$p = 4.65$ Mbar

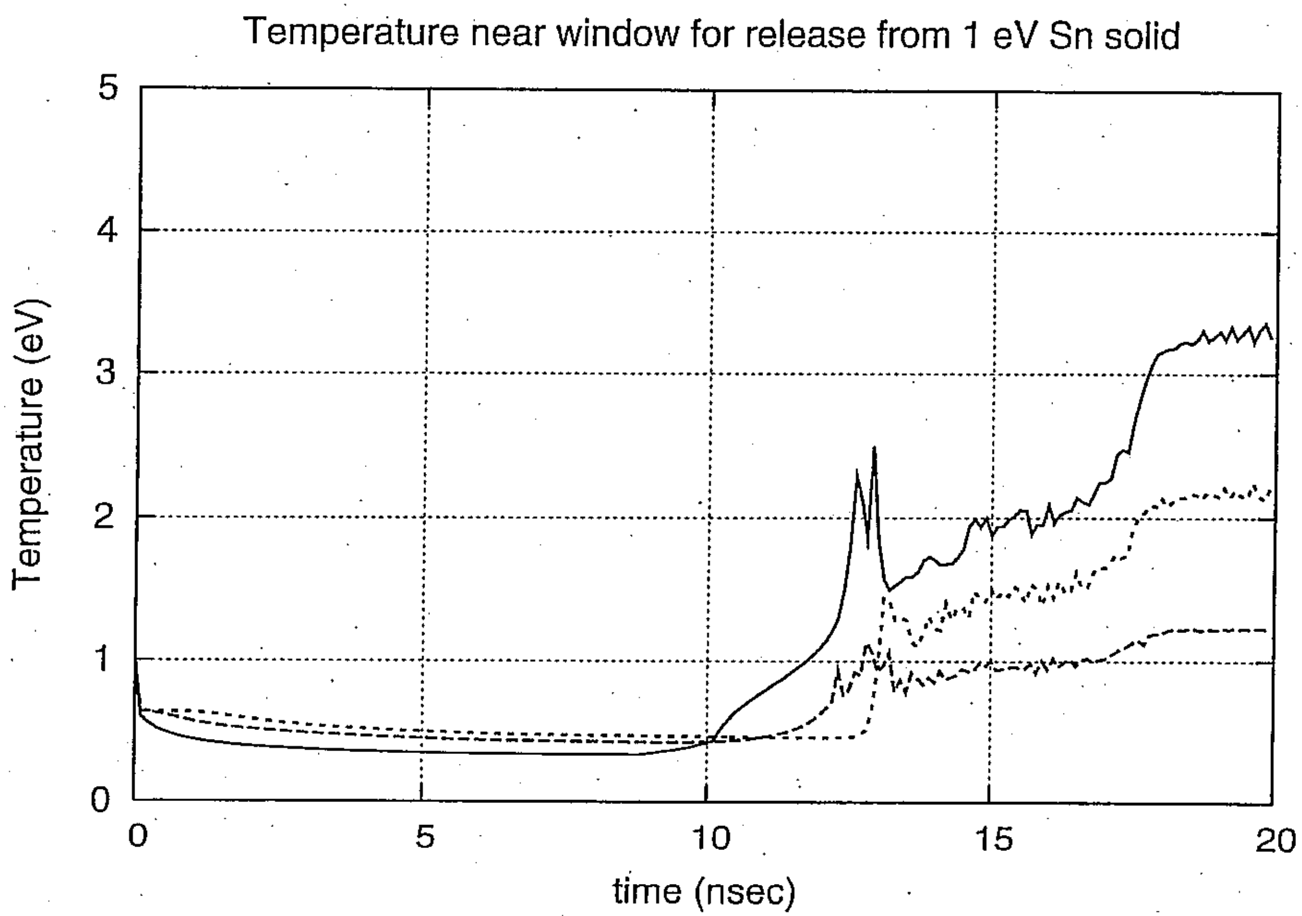




Window interaction of release from solid-density Sn (To = 1 eV)
Denisity (g/cm3)
8
7
6
5
4
3
2
1
0
5
10
15
20
Time (nsec)
-50
-40
-30
-20
-10
0
X (microns)



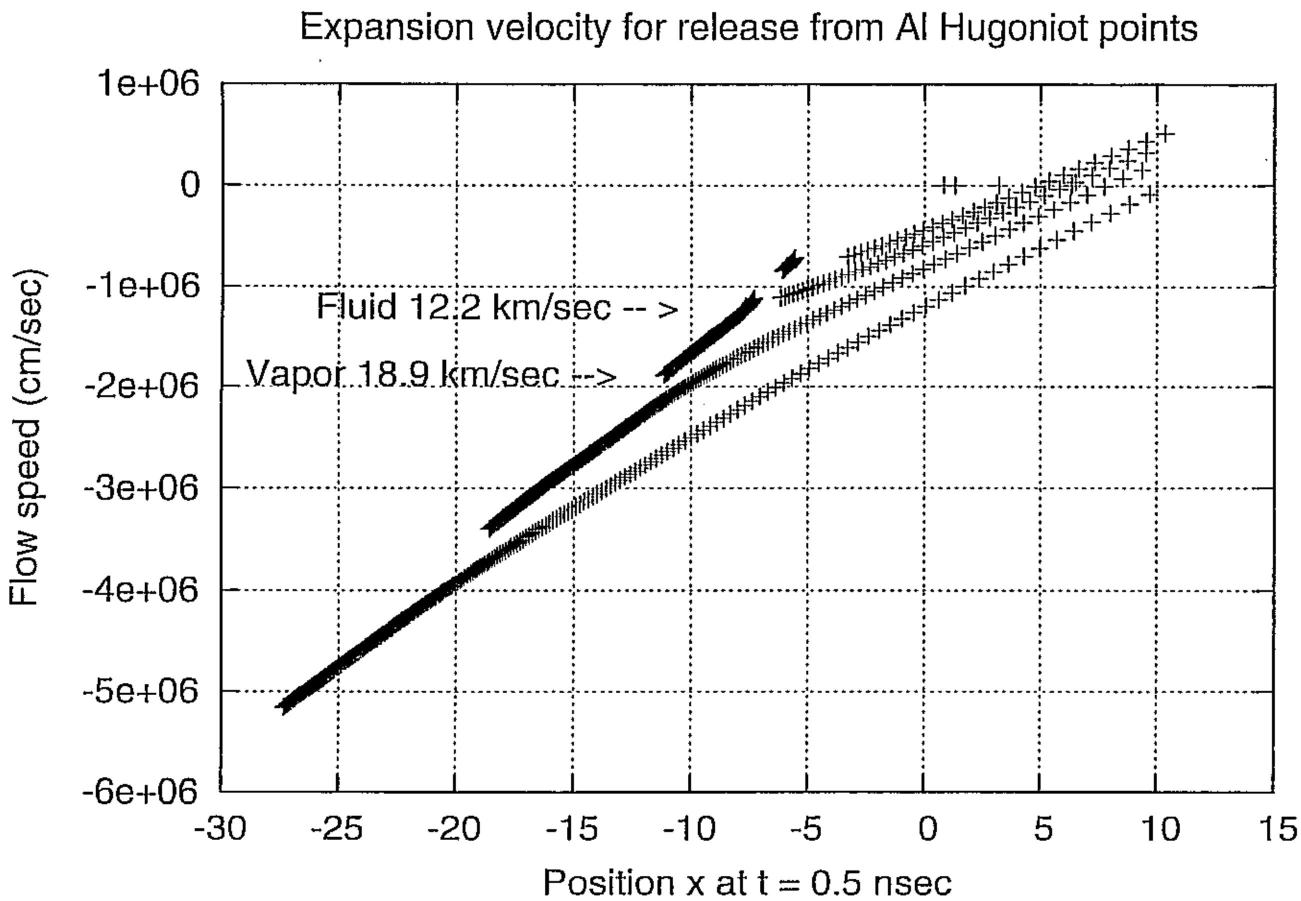
Window interaction of release from solid-density Sn (To = 1 eV)
Temperature (eV)
2
1.5
1
0.5
0
0
-10
-20
-30
-40
-50
X (microns)
0
5
10
15
20
Time (nsec)



Temperature near window for release from 1 eV Sn solid
Temperature (eV)
time (nsec)
0
1
2
3
4
5
0
5
10
15
20

Compare release speeds:

	code	+	u_p	=	v_c	v_{exptl}
FLUID	12.2 km/s	+	9.7 km/s	=	21.9	21.2 - 22.7
VAPOR	18.9 km/s	+	9.7 km/s	=	28.6	25.1 - 27.6



2-phase Hydro and Shock-Release Experiments

o Does p ~ 4.6 Mbar Al release into the 2Φ region? YES

o Does the 2Φ release wave make 2 light flashes? YES

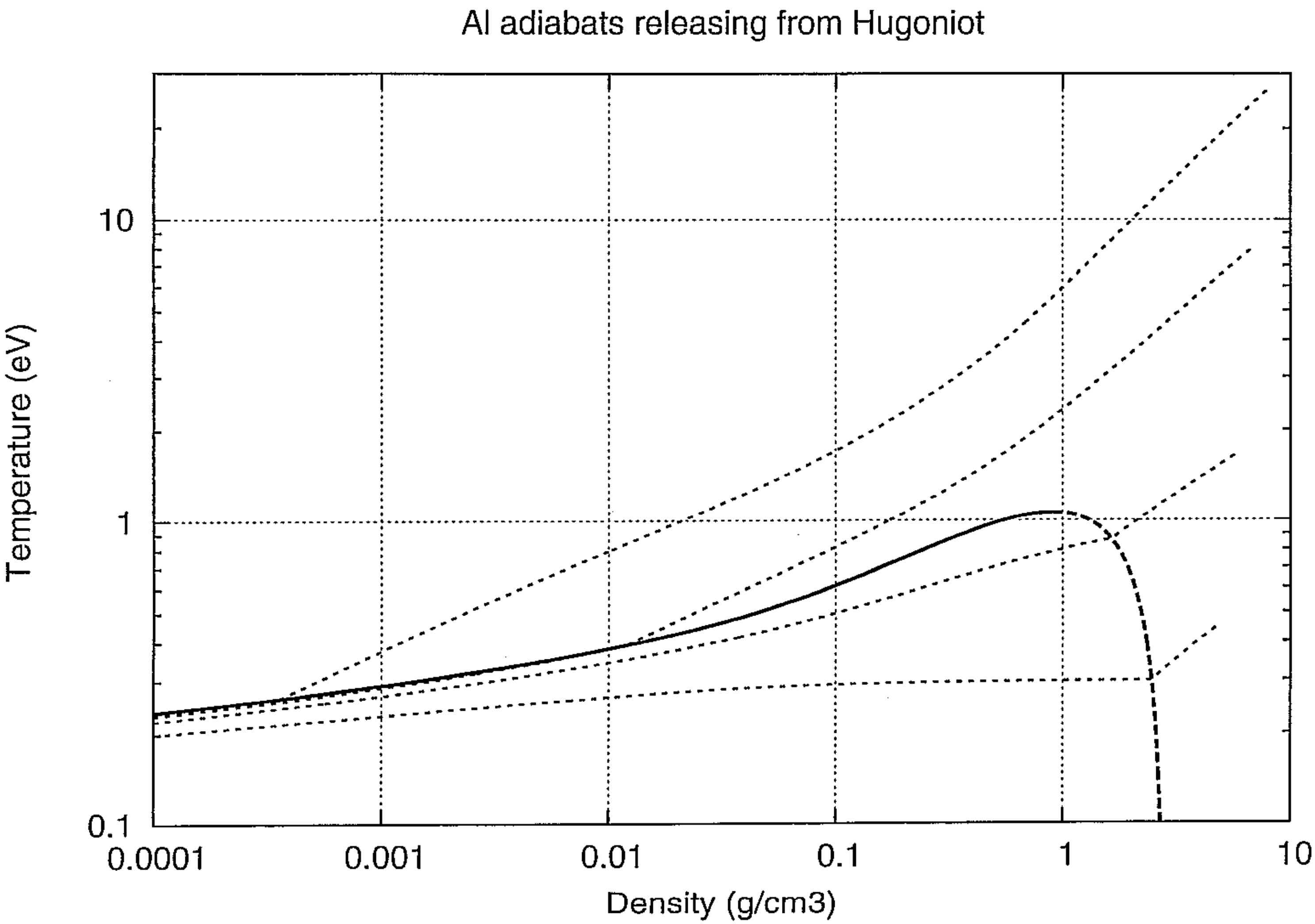
o Are speeds and times ~ correct for some aluminum EOS? YES

o Does LiF turn opaque when hit by a strong shock? Apparently

o Can we simulate the whole experiment? (shock, release, window interaction)

o Is there a clear signature in $v_1(p)$, $v_2(p)$?

The experiments constrain ρ_c, T_c and probably S_c


Al adiabats releasing from Hugoniot
Temperature (eV)
10
1
0.1
0.0001
0.001
0.01
0.1
1
10
Density (g/cm3)

NEW EOS IS BASED ON SAHA EQUATION

plus close attention to low-temperature 2-phase region

- Up to 10^9 excited states parametrized in Gj(E) ==> Gj(E, ρ)

  NIST data for neutrals

  Semiclassical SCF data for ions

- Debye, Gruneisen and Lindemann laws plus improved fluid EOS

  Cohesive energy, Bulk modulus, melting temp, latent heat, boiling point, Debye temp, Gruneisen constant, experimental molecular bonding, electron affinity, excited states

- Includes diatomic molecules and negative ions

- 2Φ EOS = Maxwell construction for liquid-vapor equilibrium

- All TD functions and accurate TD consistency

TESTED with vapor pressure, Hugoniot and QMD data

New Tungsten EOS

Low-T data: ρ_0, E_{coh}, B, T_{melt}, L_{melt}, $\Delta S_{melt} = L_m/T_m$, γ_G, Θ_D

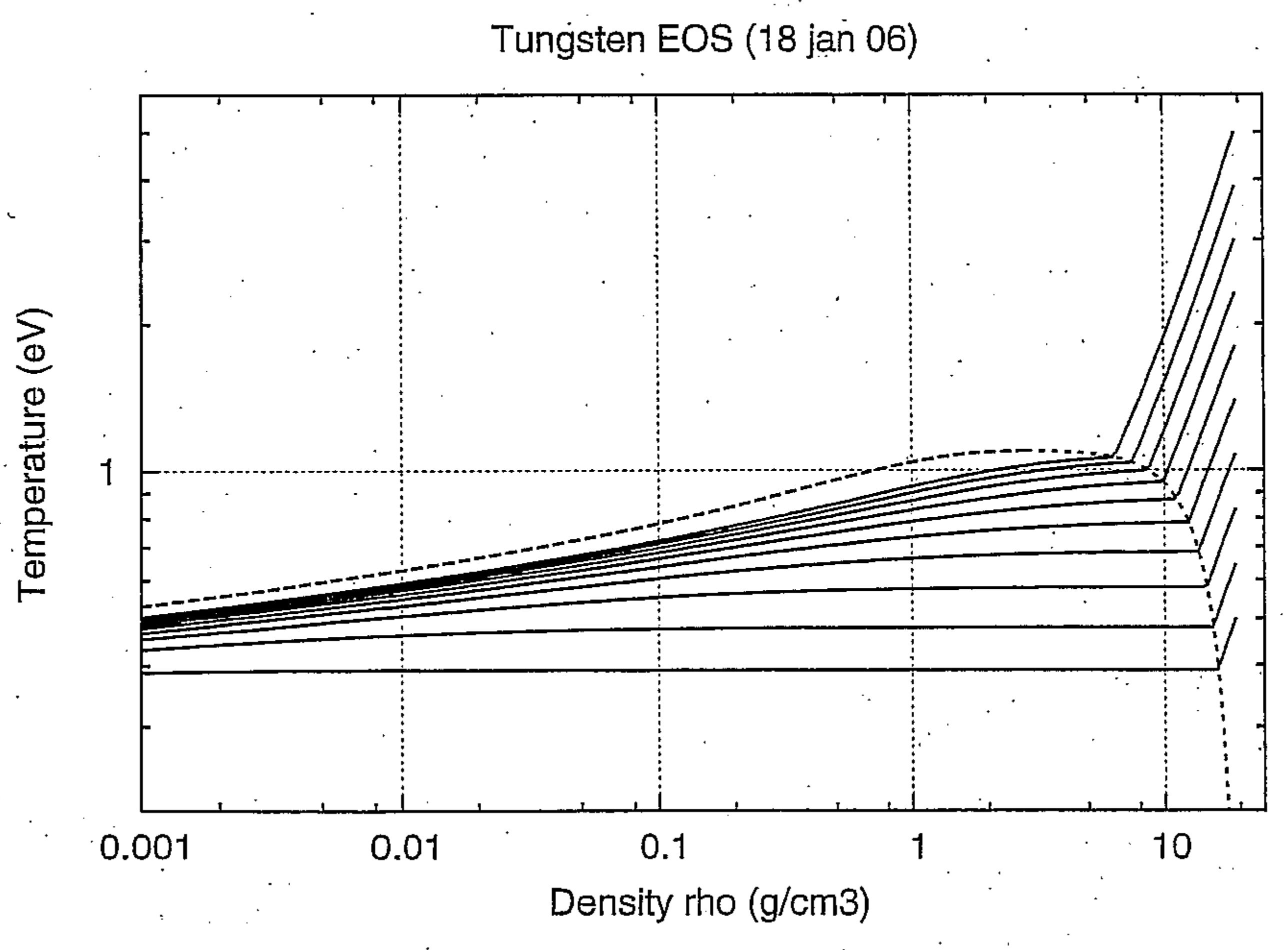
excited states of neutral atom (NIST tables)

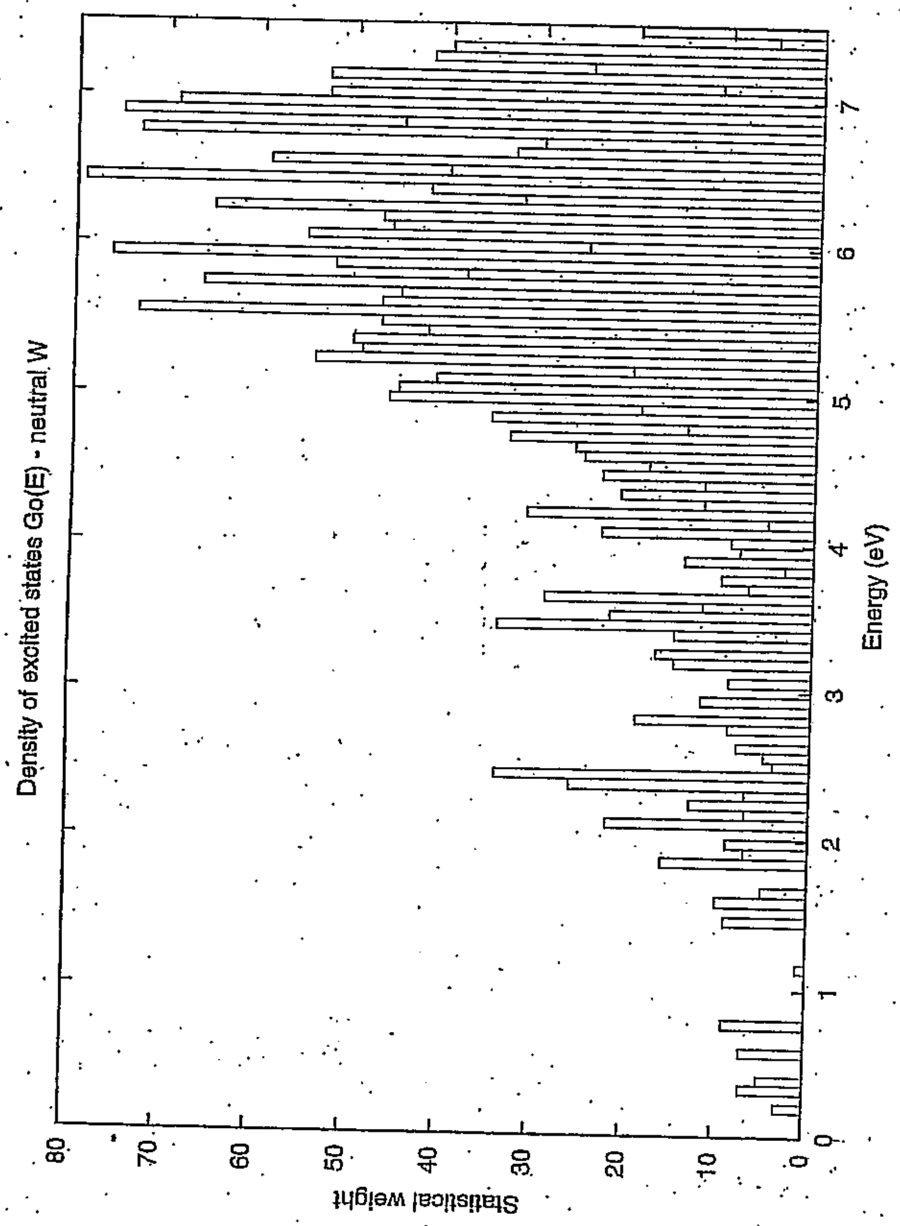
Shock-wave Hugoniot (Marsh book)

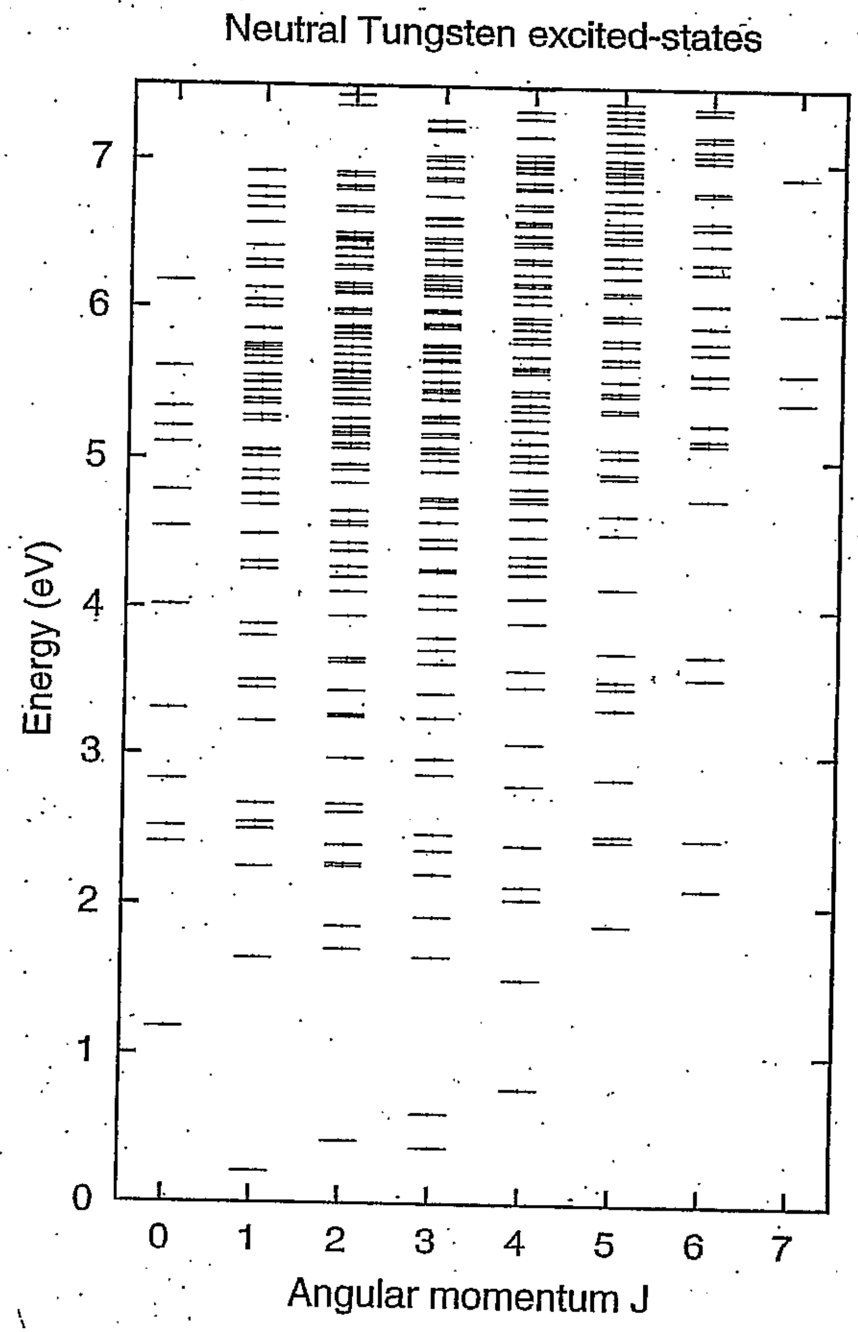
Vapor pressure (AIP Handbook)

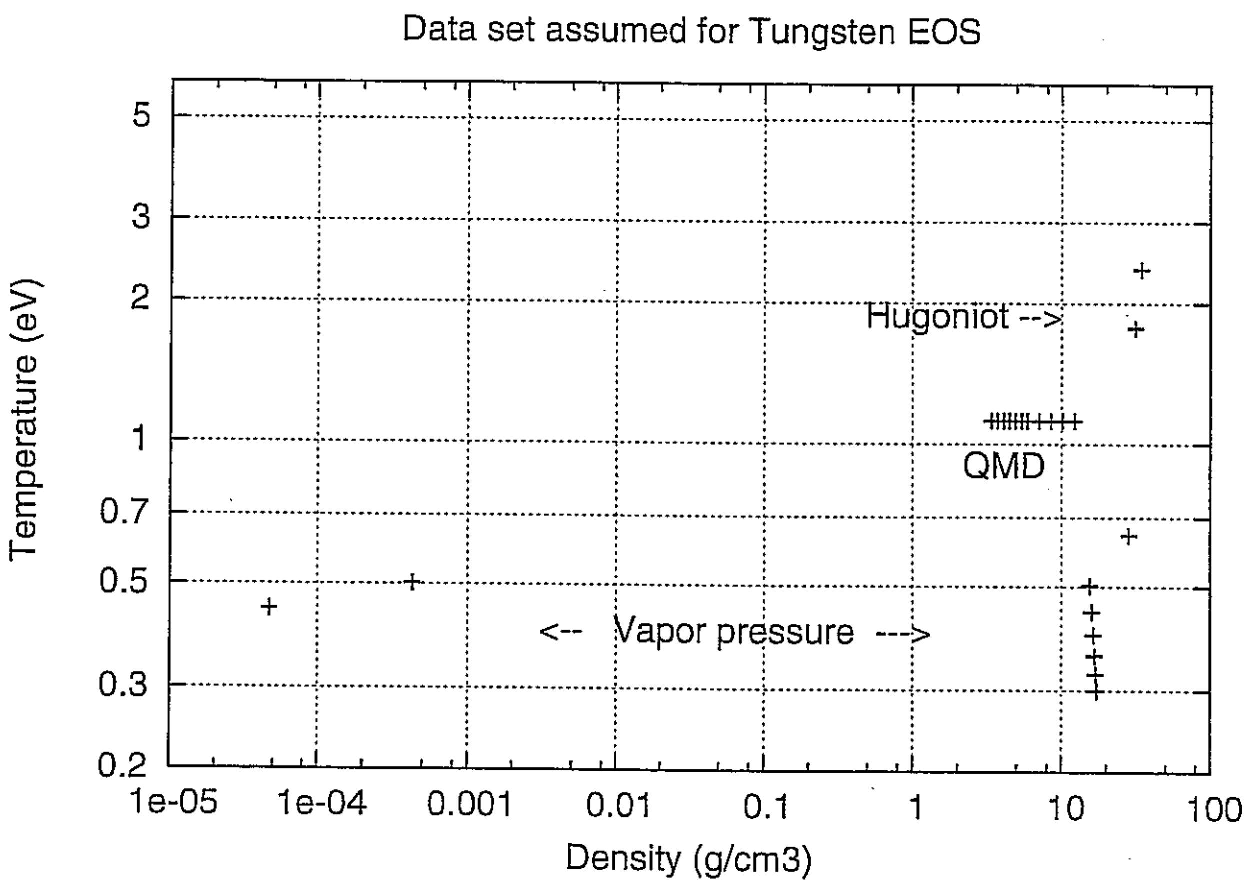
QMD calculations for T = 13000K (M. Desjarlais, SNLA)

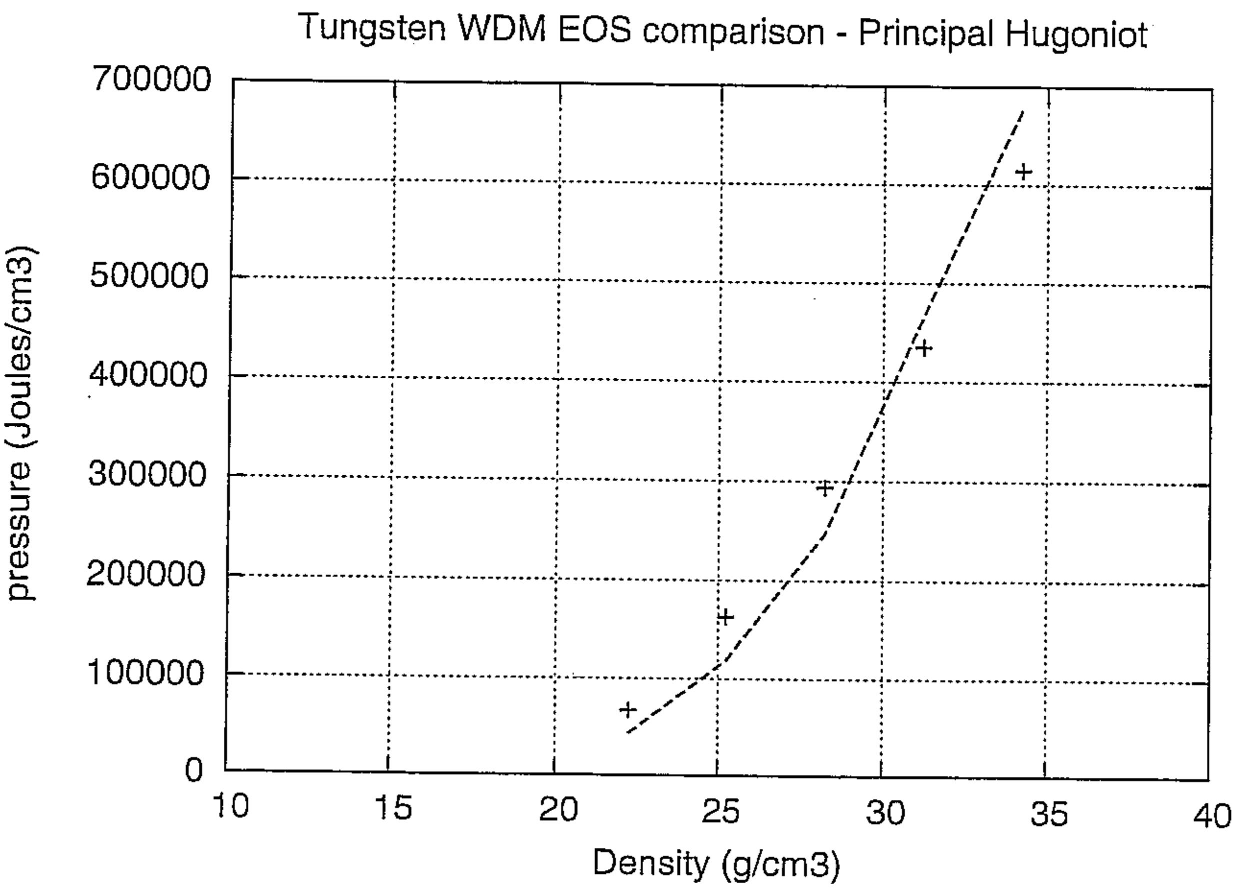
Critical point ~ 1.1 eV depends strongly on QMD data



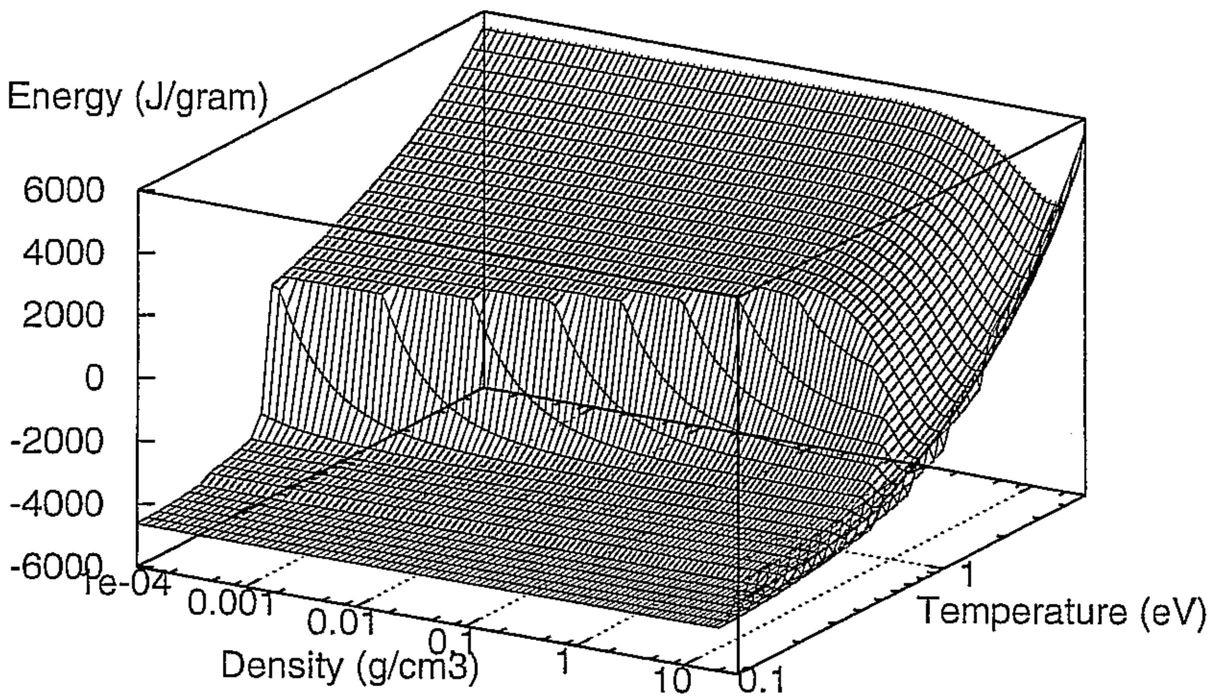

Density of excited states Go(E) - neutral W
Energy (eV)
Statistical weight


Neutral Tungsten excited-states
Energy (eV)
Angular momentum J


Data set assumed for Tungsten EOS
Temperature (eV)
5
3
2
1
0.7
0.5
0.3
0.2
1e-05
1e-04
0.001
0.01
0.1
1
10
100
Density (g/cm3)
Hugoniot -->
QMD
<-- Vapor pressure -->


Tungsten WDM EOS comparison - Principal Hugoniot
pressure (Joules/cm3)
700000
600000
500000
400000
300000
200000
100000
0
10
15
20
25
30
35
40
Density (g/cm3)

Tungsten EOS (18 jan 06)



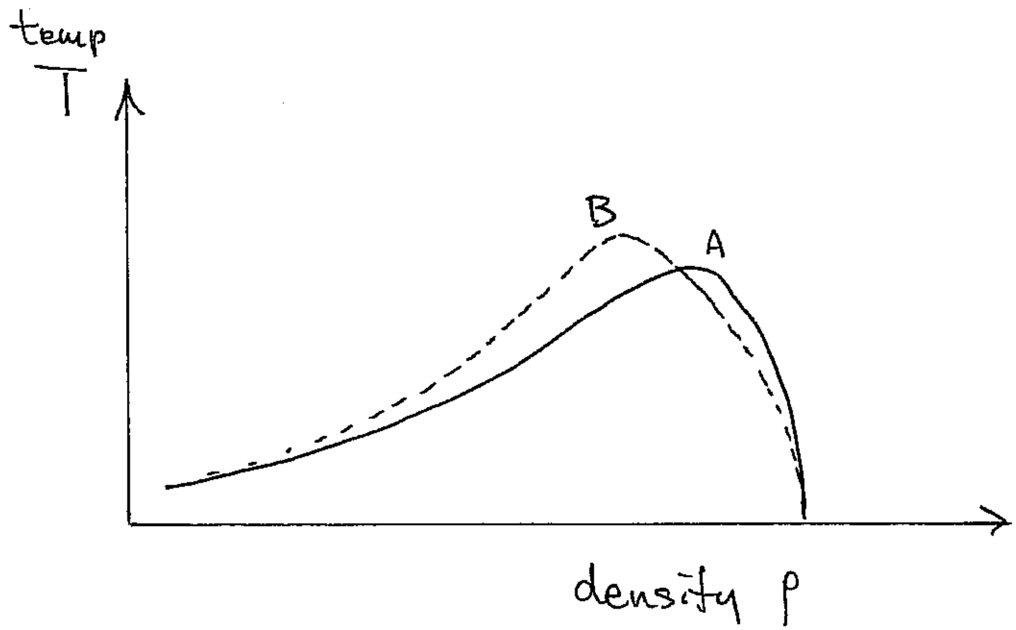
QUESTION:

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.

QUESTION: ***EOS for 2-temperature plasmas ?***

$T_e \neq T_i$ is a common type of non-equilibrium

e-i collisional heat exchange is slow

Cannot simply add electron and ion contributions to the EOS

At WDM conditions the ion p, E are ~ 50 % of total.

Should include electron-ion Coulomb interaction in Helmholtz free energy

Two-temperature EOS theory gives a formal structure

F(ρ, Te, Ti) ---> $$dF = -S_e dT_e - S_i dT_i + \frac{p}{\rho^2} d\rho$$

RMM, NATO ASI 1982

A new type of "tricritical point" with one mechanical variable and 2 T's

There are many applications of WDM information:

Contributions to fusion research

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

Radiation sources (EUV)

Cutting and machining with lasers

Plasma processing technology

Future plasma semiconductors?

Applications to geophysics, astrophysics, condensed-matter