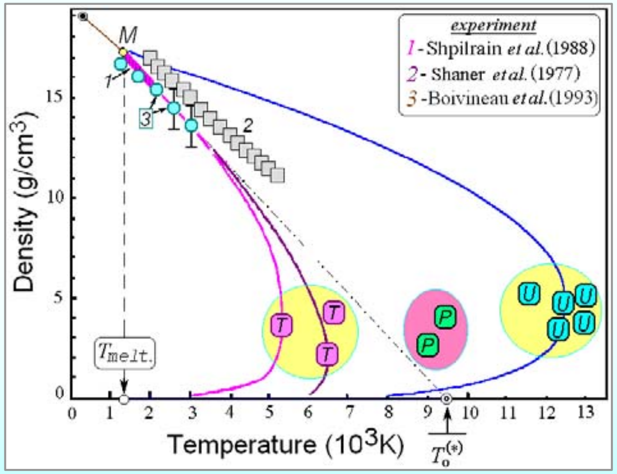
EMMI: Cosmic Matter in the Laboratory

Plasma Physics *with* **Intense Laser** *and* **Heavy Ion Beams**

3rd Workshop: Moscow, May 20-21, 2010

Heavy Ion *and* Laser Heating *in resolution of*

Uranium Critical Point Location Problem



Igor Iosilevskiy

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Moscow Institute of Physics and Technology (State University)

Support: **RAS Scientific Program** "Physics of Extreme State of Matter"
MIPT Research-Education Center "Physics of Extreme State of Matter"

Historical comments

Boris Sharkov, Meeting in ITEPh, **1997**; EMMI-Workshop, 20 May, **2010**; . . . .

! - Brilliant perspectives with HIB energy deposition, ΔE ~ 100 kJ/g. . . but now: ΔE ~ **1** kJ/g

? - What could we do with ΔE ~ 1 kJ/g *and* t_{HIB} ~ 100 ns **?**

Igor Iosilevskiy, Meeting in ITEP, **1997**

Igor Iosilevskiy, Meeting at HIF, **2002**

Igor Iosilevskiy, Meeting in GSI, **2007**

Igor Iosilevskiy, Meeting in GSI, **2009**

? ? ? Meeting in GSI, **20..??**

? - What could we do with ΔE ~ 1 kJ/g *and* t_{HIB} ~ 100 ns **?**

Study of thermophysical properties for WDM

- What to study
- How to arrange HIB energy deposition
- How to arrange measurements

HIB for thermophysical investigations

- What to study
- How to arrange HIB energy deposition
- How to arrange measurements

Basic point

- Careful choice of investigated substance and physical problem

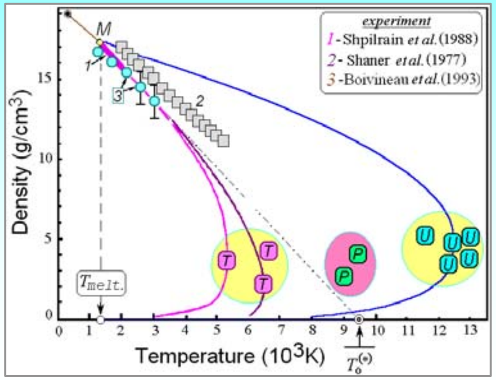
Criteria

- great uncertainty
- great applied importance
- fundamental physics

? - What substance - ? What property - ? What parameters - ?

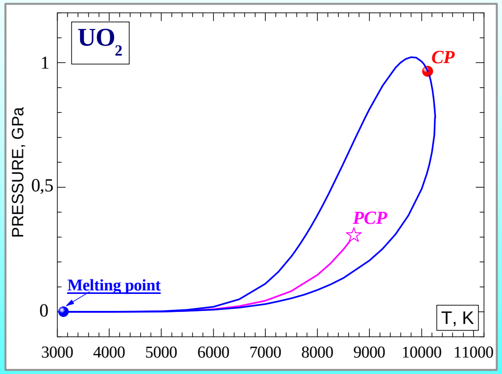
Low energy deposition – what could we study via HIB ?

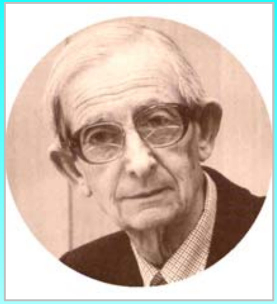
Two outstanding goals



- **Uranium Critical Point Location Problem**
  - applied importance
  - phenomenology
  - fundamental physical problem

- **Non-congruent Phase Transitions in High Energy Density Matter**
  (*uranium-bearing fuels* (UO_2, UC, UN ...) *and other mixed substances*)
  - applied importance
  - phenomenology
  - fundamental physical problem





«Extreme State of Matter»

Int. Conference "Equation of State"

Russia, Elbrus, 1990 -2010

Int. Conference "Khariton's Science Readings"

Russian Federal Nuclear Center, Sarov, 2005 -2010

Int. Conference "Zababakhin's Science Readings"

Russian Federal Nuclear Center, Snezhinsk, 2005 -2010

ЗАБАБАХИНСКИЕ НАУЧНЫЕ ЧТЕНИЯ

EQUATIONS OF STATE FOR MATTER
XXV INTERNATIONAL CONFERENCE
ELBRUS 2010

Uranium Critical Point Location Problem

- Applied importance
- Phenomenology
- Fundamental physical problem

In cooperation with:

Victor Gryaznov (*IPCP RAS) and* Artem Ukrainets & Katya Romadinova (*MIPT)*

Uranium Critical Point Location Problem

Uncertainty

Recommendations of IVTAN-Database *(1982)*

Critical Point Parameters

Cr	9620	968	0.023
Mo	11150	546	0.0365
W	13400	337	0.043
V	12500	1078	0.027
Nb	19040	1252	0.030
Ta	20570	13500	0.036
Ti	11790	763	0.037
Zr	16250	752	0.051
Hf	18270	938	0.046
Sc	8350	408	0.048
Y	10800	374	0.068
La	11060	335	0.078
Th	14950	488	0.072
U	11630	611	0.045
UO_2	7560	122.6	0.163
UF_6	504.5	4.59	0.255
Pu	10000	324.2	0.081
PuO_2	7620	101	0.202
Li	3680	60	0.055
Na	2503	25.64	0.111
NaCl	3400	35	0.266
K	2280	15.8	0.202
KCl	3200	22	0.415
Rb	2106	13.22	0.246
Cs	2043	11.75	0.308

Discussion: *I. Iosilevskiy & V. Gryaznov, Journ. of Nuclear Materials,* **344** (2005)

Critical Point Parameters

Recommendations of IPCP RAS-GSI Database

	Pressure P_c, kbar	Temperatue T_c, K	Density ϱ_c, g/ñm^3	Entropy S_c, J/g/K
Be	2.87	8877	0.398	13.18
Mg	2.46	3957	0.553	3.789
Na	0.47	2473	0.240	3.281
Zr	9.88	14860	1.634	1.693
Hf	11.74	15810	3.610	0.885
V	9.19	9915	1.631	2.718
Nb	11.06	19180	1.701	2.023
Ta	9.93	13530	4.263	0.923
Cr	9.91	7797	2.660	2.332
Mo	7.59	10180	3.690	1.520
W	11.80	15750	4.854	0.837
Fe	11.31	8787	2.183	2.496
Co	5.55	9157	1.890	2.458
Ni	10.42	7547	2.092	2.518
Zn	3.28	3079	2.381	1.468
Cd	0.87	2510	2.283	0.840
Ag	10.64	7053	3.279	1.118
Au	6.14	8515	6.061	0.624
Re	15.91	18710	6.024	0.824
Ir	13.40	16220	6.061	0.780
Pt	6.21	11430	5.236	0.807
Sn	2.39	8175	1.592	1.123
Bi(VI)	2.25	4869	2.937	0.529
U	7.70	9637	4.505	0.727

After
D. Varentsov
FAIR-Russia School
Moscow, 2009

Uranium Critical Point Location

fifty years old problem

List of Uranium CP parameters estimations

$T_C \approx$ 12'000 K (*the early estimation*) Braut (**1957**)

T_c [K]	p_c, bar	ρ_c g/cm^3	Z_c	ρ_s/ρ_c (*)	References
6'618	4160	4.12	0.437	4.60	Young D. (1977)
7'533	**798**	1.03	0.295	**18.4**	Gates D. et al. (1960)
6'200–7'663	-	-	-	-	Goldstein R.(1989) Hess H.(1995)
8'317–9'112	-	-	-	-	Guldberg C. (1890).
8'730	2360	5.17	**0.150**	3.67	Martynyuk M. (1989).
9'000 (z_c = 3)	5000	2.60	0.6	7.42	Likalter A. (1997)
9'400 (z_c = 3)	6000	2.59	**0.706**	7.32	**Likalter** A. (1985 - 1996)
11'630	6110	5.30	0.284	**3.58**	**Fortov** V. *et al.* (1975)
11'679–12'995	-	-	-	-	Kopp I. (1967) Lang G. (1977)
12'400	4800	3.55	0.312	5.34	Morris E. (1964)
12'434	4950	3.78	0.302	5.02	Gathers-Shaner-Young (1974)
12'500	-	-	-	-	Grosse A. (1961)
13'034	5136	4.03	0.280	4.71	Hornung K. (1975)
13'043	**8'487**	5.17	0.361	3.66	Young D. & Alder B. (1971)

Table from:
H. Hess, H.Schneidenbach
Z. Metallkd. (***2001***)
Vapor Pressure and Critical Data for Uranium

9'636	**7'700**	4,50	Wide-range EOS	**Bushman - Lomonosov**
7'000	**1'712**	3,30	Extrapolation of Liquid $\rho(T)$	Apfelbaum – Vorob'ev (**2009**)
5'500– 6'500	100 – 1'000		(*estimation* – **thermal EOS** calibration))	**Iosilevskiy** (**1990**)
6'840 **12'800**	**4'440** **8'450**		(Plasma model – **thermal** calibration) (Plasma model – **caloric** calibration)	**Iosilevskiy & Gryaznov** *SAHA-code, JNM,* (**2005**)

Additions IL

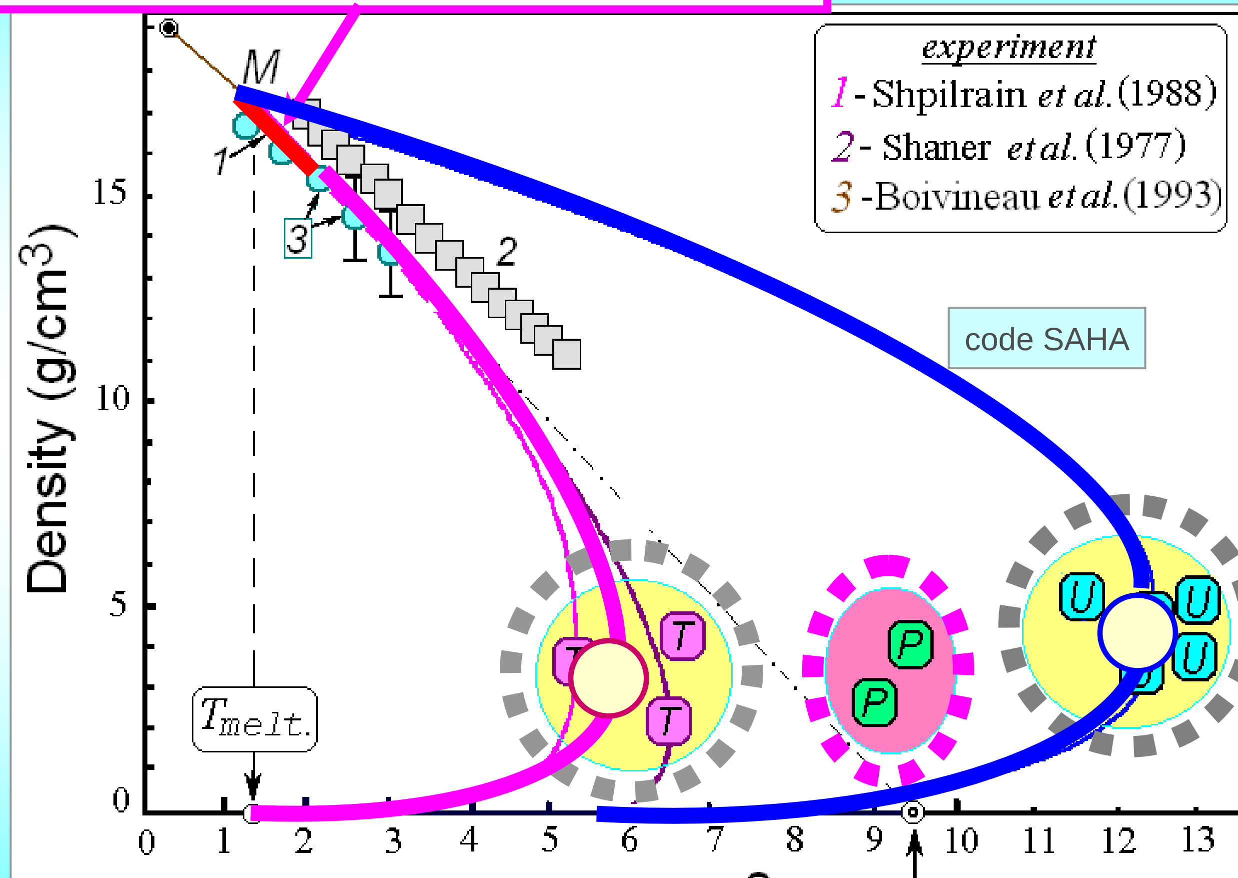
Uncertainty in high-temperature density-temperature diagram ... ical point

Shpilrain *et al.* (1988)
Static experiment: - ρ_{liquid} (1400 < T < 2100 K)
high accuracy – 0.5 %

Thermal variants (T)
Yound & Shaner (1977)
Gathers et al. (1986)
Iosilevskiy (1991)
Iosilevskiy & Gryaznov (**SAHA-T**)
- - - - - - - - - - - - - - - -
Correlation
T_C ⇔ Thermal expansion

Caloric variants (U)
Brout (1957)
Grosse (1961)
Morris (1964)
Yound & Alder (1971)
Gathers et al. (1974)
Fortov et al. (1975)
Hornung (1975).....
Iosilevskiy & Gryaznov (code **SAHA-E**)
- - - - - - - - - - - - - - - -
Correlation
T_C ⇔ Vaporization heat

Plasma Hypothesis (P)
Likalter (1981)
Likalter + Hess (1997)
- - - - - - - - - - - - - - - -
Correlation
T_C ⇔ Ionization Potential

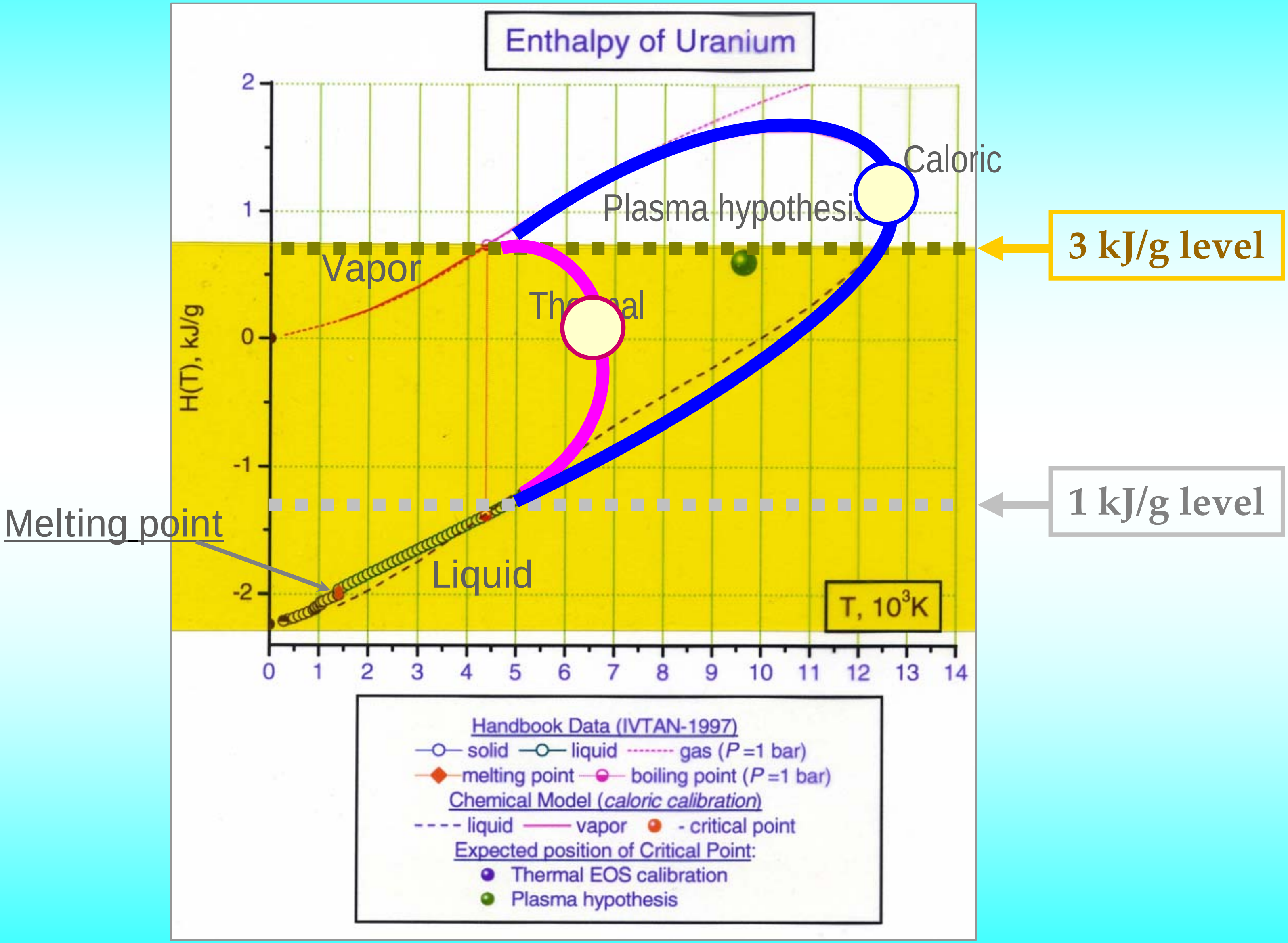


Extrapolation: (*i*) Guggenheim's formula, (*ii*) Law of Correspondent States (*Iosilevskiy, 1990*), (*iii*) SAHA-code (*Iosilevskiy & Gryaznov, 2005*)

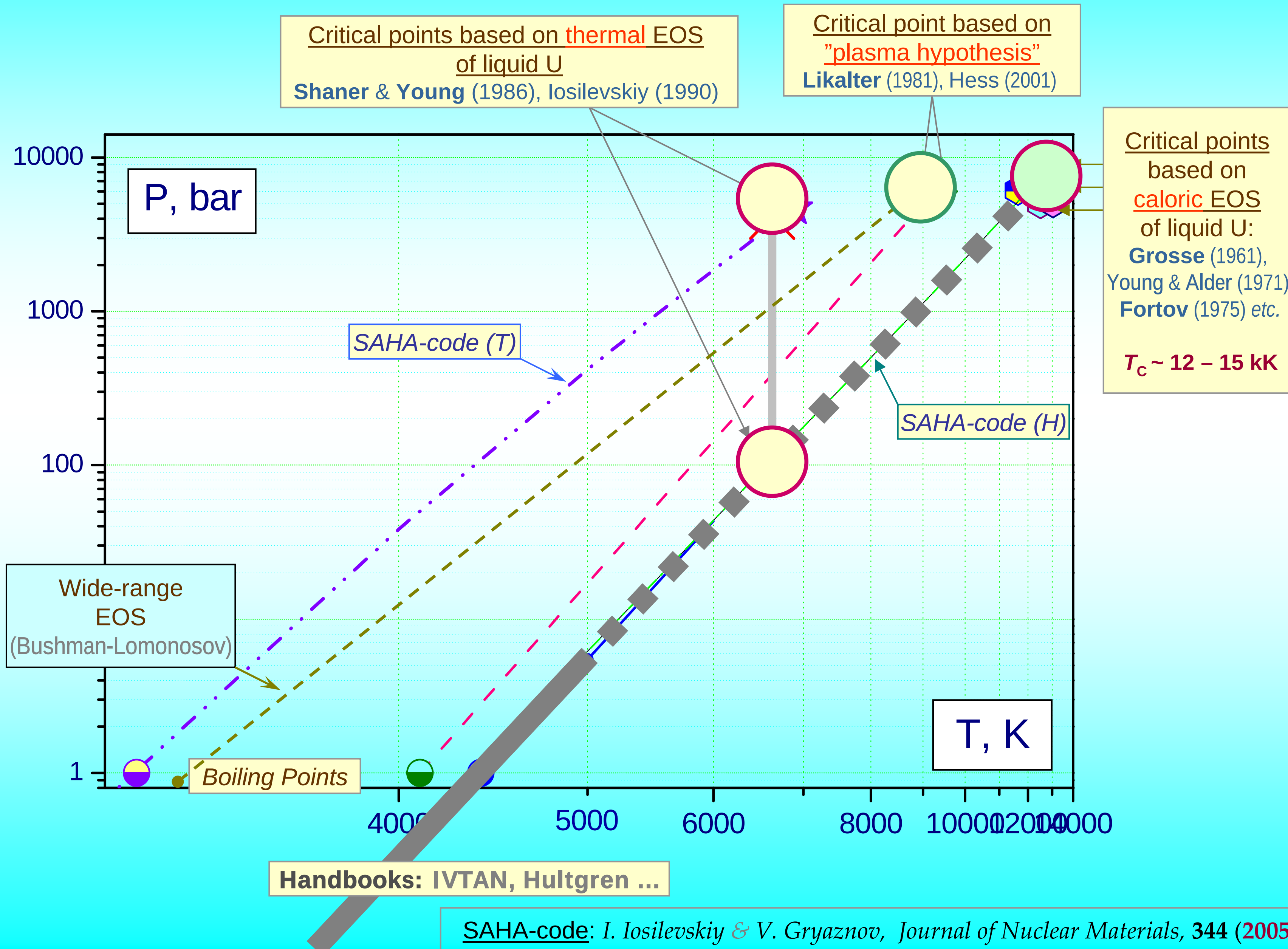
(*i*) $$\frac{\rho_{l,v}}{\rho_c} = 1 + b_1\left(1 - \frac{T}{T_c}\right) \pm b_2\left(1 - \frac{T}{T_c}\right)^{\beta}$$

(*ii*) $$\left(\frac{\rho(T/T_{cr})}{\rho_{cr}}\right)_U = \left(\frac{\rho(T/T_{cr})}{\rho_{cr}}\right)_{Cs}$$

Uncertainty in high-temperature caloric phase diagram



Uncertainty in Uranium Critical Pressure



SAHA-code: *I. Iosilevskiy & V. Gryaznov, Journal of Nuclear Materials*, **344** (2005)

The Problem:

Experimental data:

Liquid density $\rho(T)$ - (T < 5 kK)
Liquid enthalpy $H(T)$ - (T < 5 kK)
Vapor pressure $P_s(T)$ - (T < 5 kK)

Semi-empirical rules:

Convexity of liquid density $\rho_L(T)$
Quasi-linear vapor pressure $\ln P_s(T^{-1})$
Universal evaporation enthalpy $\Delta H(T)$

Incompatible

Dilemma:

Access semi-empirical rules
and
Deny experimental data

or

Access experimental data
and
Violate semi-empirical rules

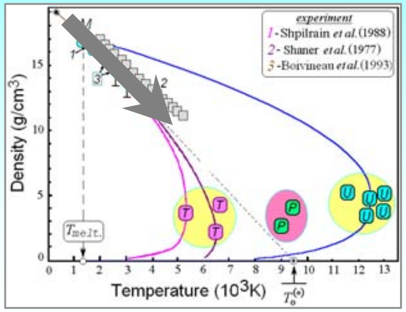
Hypothetical resolution ?

Discussion: *I. Iosilevskiy* (Elbrus-1990) // *I. Iosilevskiy & V. Gryaznov, Journal of Nuclear Materials,* **344** (2005)

In Search *of* Resolution *for*
Uranium Critical Point Location Problem

Low temperature — *High temperature*

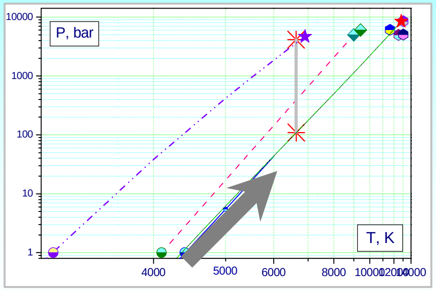
? Wrong experiment ⇔ wrong extrapolation ?



liquid density $\rho(T)$ - **?**

liquid enthalpy $H(T)$ - **?**

vapor pressure $P_s(T)$ - **?**



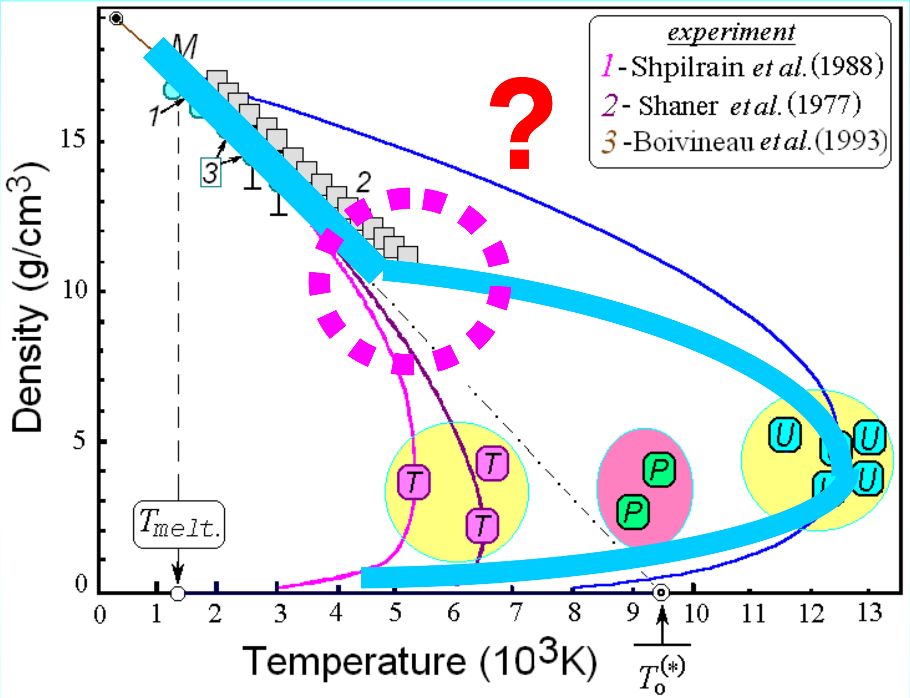
! unbelievable !

We have enough reason to expect

Violation of semi-empirical rule(s)

Hypothetical violation of semi-empirical rule(s)

Lost *of* convexity *for* $\rho_L(T)$?



? What physical reason can approve this violation ?

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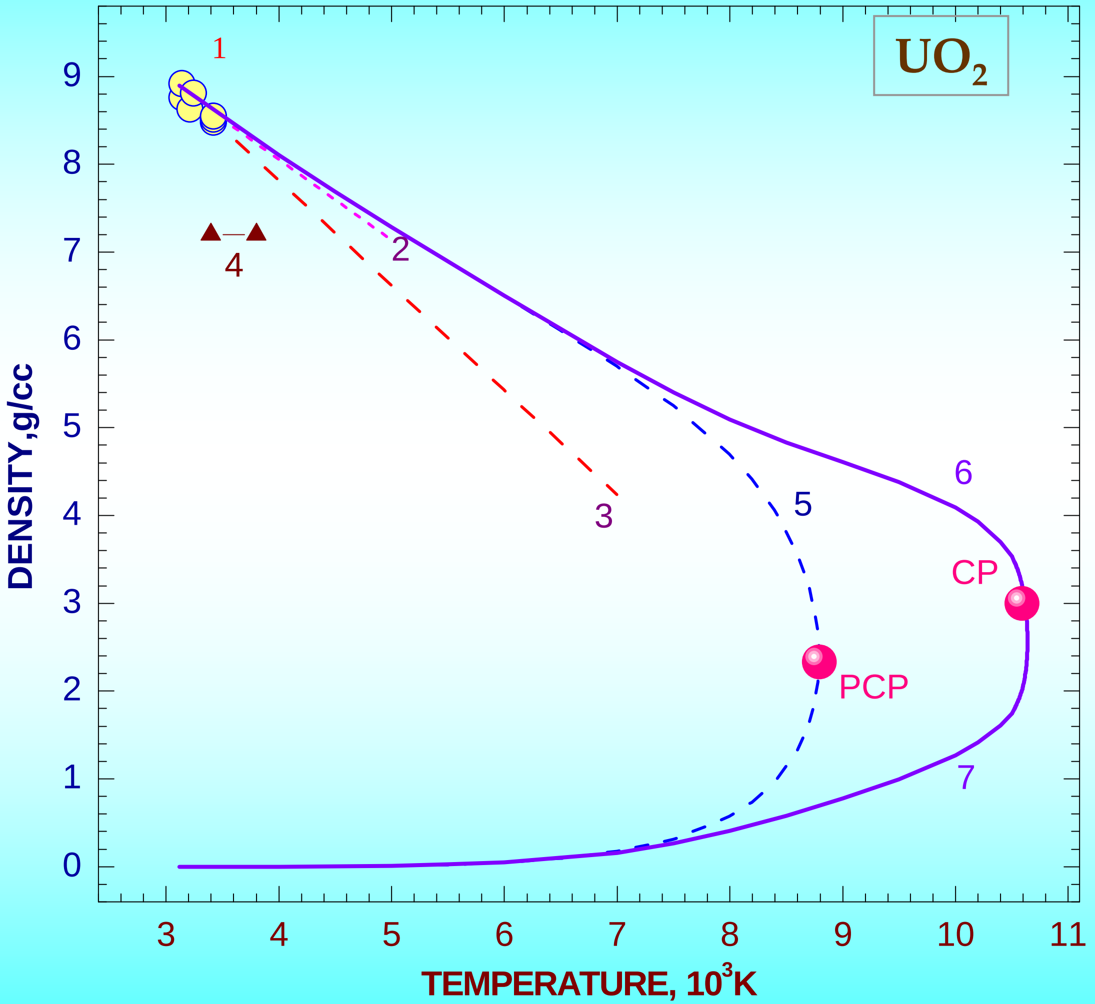
Elbrus_1990-2010

Exception

Non-congruent evaporation !



Iosilevskiy I, *Hyland G., Ronchi C., Yakub E. "An Advanced Equation of State of UO2 up to the Critical Point"* - *Trans. Amer. Nucl. Soc*. **81** 122 (1999)// - *Int. Journ. Thermophys.* **22,** 1253 (2001)// *Contrib. Plasma Phys.* **43**, (2003)

What physical reason can approve the lost of convexity for $\rho(T)$ two-phase boundary in uranium ?

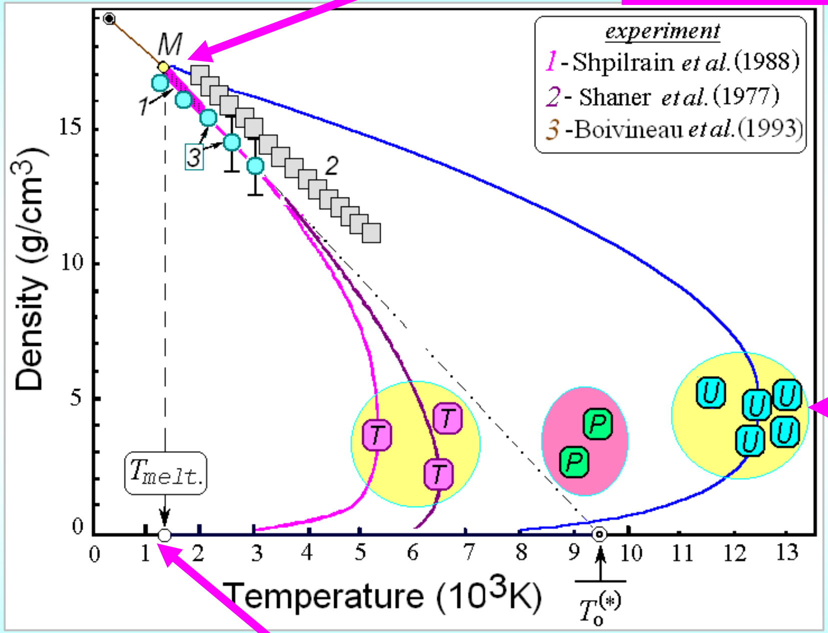
${}_{92}U^{238}\ \{5f^3\ 6d\ 7s^2\}$

UO_3

Z = 3

$U^{+3} + 3e^-$

$\Gamma_D \approx 3300\ //\ n_e \lambda_e^3 \approx 900$



$U^{+1} + e^-$

$\Gamma_D \approx 80\ //\ n_e \lambda_e^3 \approx 10$

Z = 1

Z = 0

NB !

Drastic ***change*** *of* ***effective ion-ionic interaction*** during thermal expansion of liquid uranium and decrease of electronic degeneracy

Drastic ***change*** *of* ***phase behavior*** of evaporating uranium (ρ-*T diagram)*

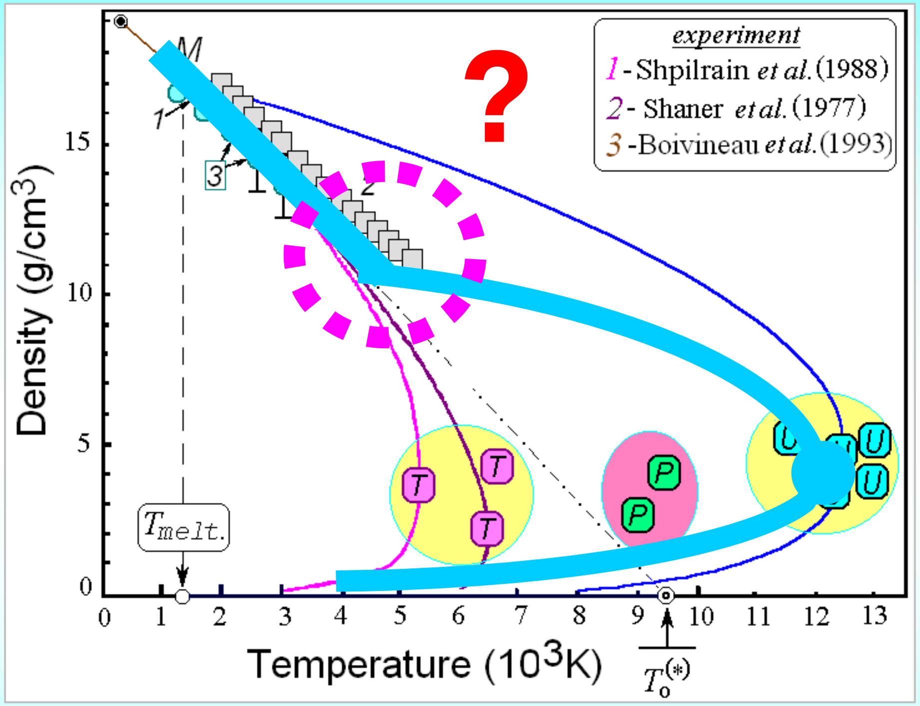
Hypothetical resolution - I

Discussion:

I. Iosilevskiy & Int. Conference: *Subsecond Thermophysics, Moscow,* 2008// *FAIR-Russia School, Moscow,* 2009

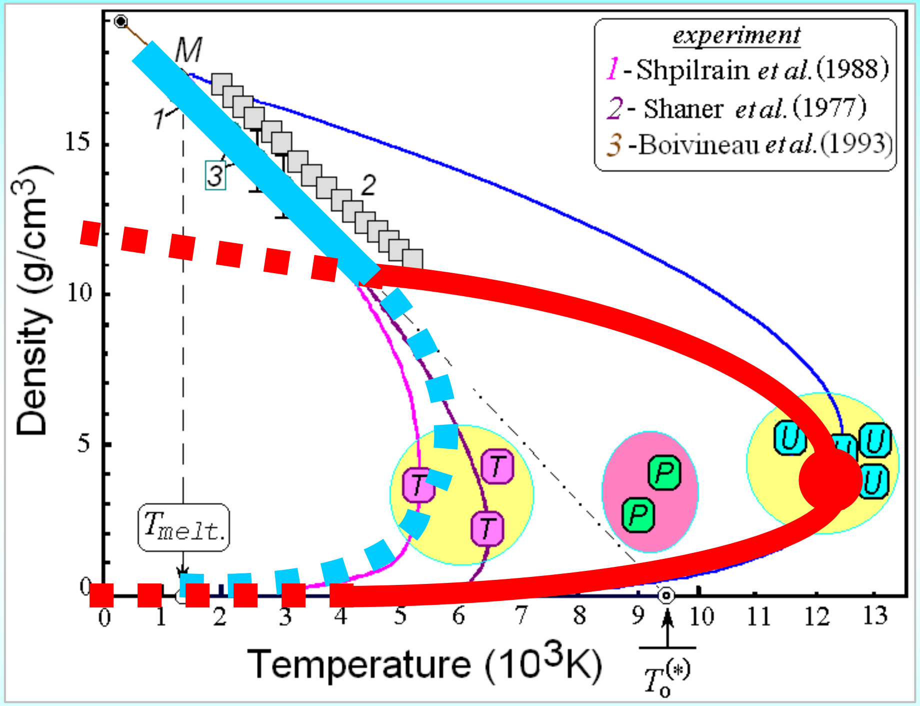
Hypothetical resolution of uranium critical point location problem

? ρ *-T* phase boundary consists of **TWO** fragments **?**



Hypothetical resolution of uranium critical point location problem

? ρ *-T* phase boundary consists of **TWO** fragments **?**



«high-density phase» ⇔ «low-density phase»

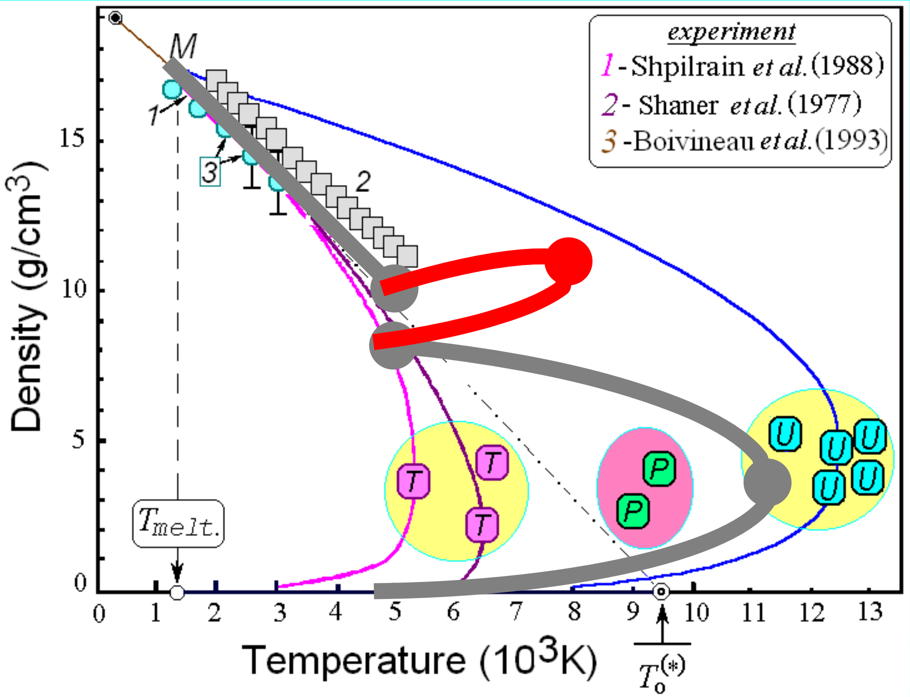
Hypothetical resolution - II

Transition from the "high-density phase" to the "low-density phase" must not be continuous

Additional phase transition ?

Discussion: *Landau & Zel'dovich* (1944) // *Norman & Starostin* (1970) // . . . .

Additional phase transition ?



Very exotic but not fantastic !

! Only 1 – 2 kJ/g is needed !

Fundamental Physical Problem

Phase transition in a system

with

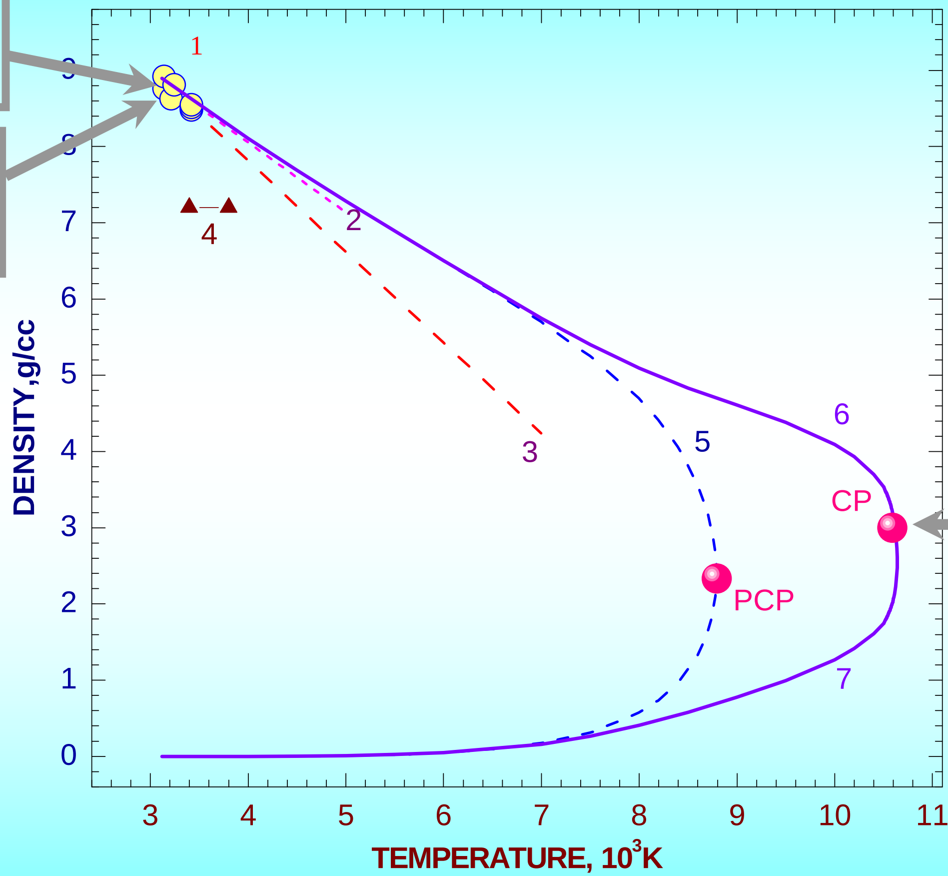
varying mean-particle interaction

Non-congruent evaporation in UO_2 ⇔ the same physical problem

Phase transition in the system with varying composition

Molecular model
UO_2 ⇔ UO_2

Ionic model
U^{+4} ⇔ O^{-2}



Ion-molecular mixture
U ⇔ O ⇔ UO
UO^+ ⇔ UO_2^+
O^- ⇔ UO_2^- ⇔ UO_3^-

Phase transition in the system
with **varying mean-particle interaction !**

Fundamental Physical Problem

What could we do ?

- **Study via simplified analytical plasma models**

  One-component plasma model on *uniformly-compressible* compensating background {OCP(~)}

- **Study via direct numerical simulation**

  FT-DFT_MD // Monte-Carlo // . . . .

- **Experimental study:**

  Exploding wires, ... *etc*

  Heavy Ion Beam

  Surface Laser Heating

HIB for thermophysical investigations

– How to arrange HIB energy deposition

Priorities

- Uniformity of heated material
- Thermodynamic equilibrium

– How to arrange measurements

Priorities

- Direct measurement of thermodynamic parameters
  without intermediate hydrodynamic re-calculations
- Energy deposition control

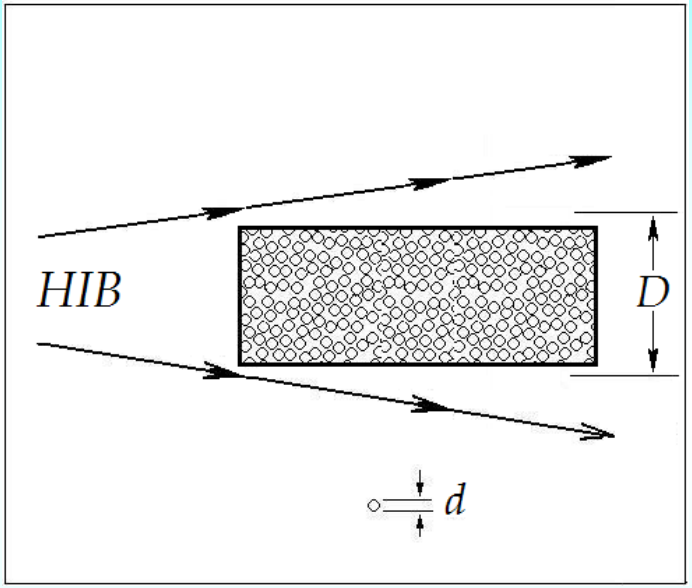
HIB heating of highly dispersive materials –
– very promising for thermophysical investigations (*)

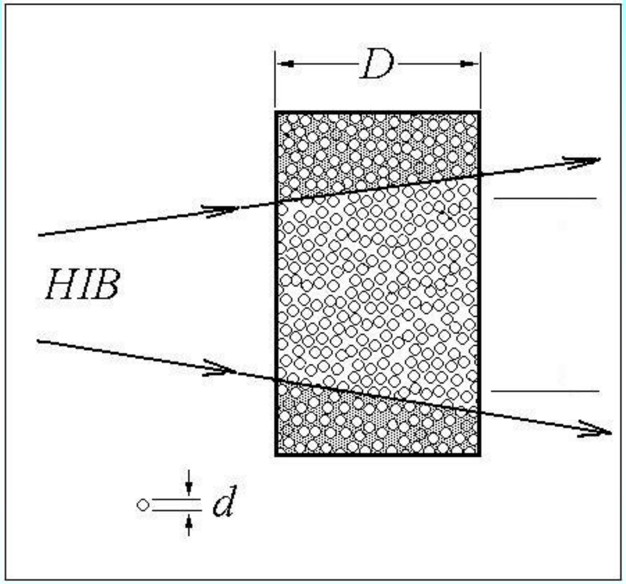
* Iosilevskiy I. // Int. Conf. *Intense Ion Beam Interaction with Ionized Matter* // Moscow, ITEP Publishing (**1999**)

* Iosilevskiy I., Gryaznov V. // XIV Int. Conf. *Heavy Ion & Inertial Fusion* // Moscow, ITEP Publishing (**2002**)

How to arrange HIB energy deposition

HIB heating of highly dispersive porous materials –
– very promising for thermophysical investigations (*)





Advantages:

– uniform quasi-free equilibrium expansion of each grain
– no fast hydrodynamic movement
– surface thermodynamic parameters are equal to the bulk ones
– porosity (ρ_{00}/ρ_0) is well-controlled parameter

* Iosilevskiy I. // Int. Conf. *Intense Ion Beam Interaction with Ionized Matter* // Moscow, ITEP Publishing (**1999**)

* Iosilevskiy I., Gryaznov V. // XIV Int. Conf. *Heavy Ion & Inertial Fusion* // Moscow, ITEP Publishing (**2002**)

Moment “X”

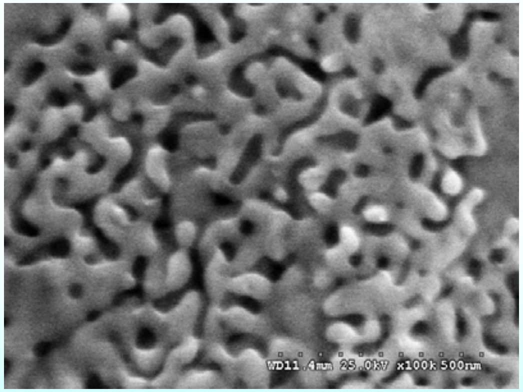
* * * * * * * * * * *

Basic idea

<u>Critical event</u> – Exhausting of free volume for grain’s thermal expansion

!! **<u>In this moment we obtain:</u>**

- **Uniform** and **homogeneous** state of investigated material
- **Known density** (*due to porosity and initial density control*)



|← 1 µm →|

!! **<u>It means :</u>**

- **End** of free quasi-**isobaric** expansion
- Fast **increasing** of bulk **pressure**
- **Start** of stressed quasi-**isochoric** expansion

Pressure Jump

Moment "X"

* * * * * * * * * * *

Pressure Jump

!! **<u>If we catch this moment</u>**

<u>and if we know</u>:

- temperature (*surface*)
- energy deposition (*beam control*)
- density (*porosity control*)

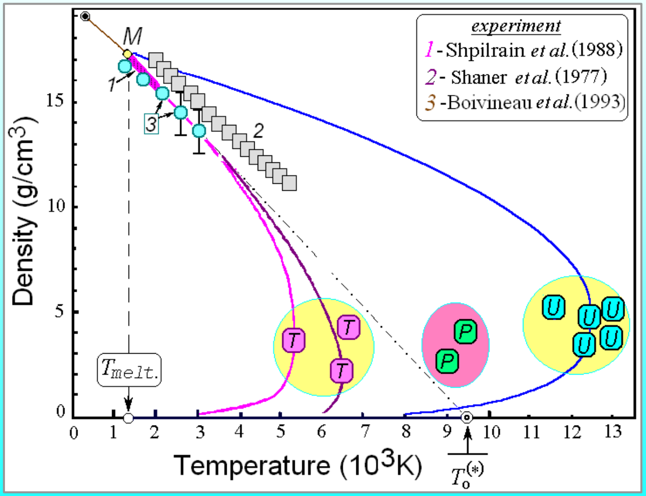
!! **<u>We obtain</u>**:

- Density of expanded liquid $\rho(T)_{liquid}$ (*or* $\rho(H)_{liquid}$)
- Thermal (*or caloric*) expansion coefficient

$$\alpha_P = (\partial\rho/\partial T)_P \qquad \alpha_P{}^* = (\partial\rho/\partial U)_P$$

* * * * * * * *

- Heat capacity $C_P = (\partial U/\partial T)_P$



!! **<u>Hypothetically</u>**:

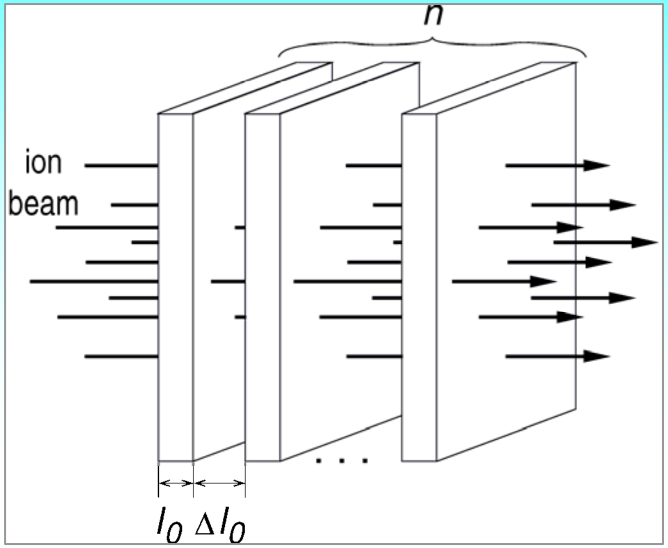
- sound speed,
- vapor pressure,
- electro-conductivity … *etc.*

Quasi-static heating of a stack target

***$t < t_x$:* quasi-static heating of a stack of foils**

- ρ, P, ε (and T) are spatially uniform $\quad u_l = \frac{1}{2}\alpha\, l\, q$
- expansion velocity

  l – foil thickness, q – heating rate, $\quad \alpha = \left(\frac{\partial P}{\partial \varepsilon}\right)\left(\frac{\partial P}{\partial \ln \rho}\right)^{-1}$
- kinetic energy $E_{kin} << \varepsilon$



Thermal (not hydrodynamic) expansion if the foils are thin:
$\Delta\rho/\rho << 1$ over $t_s = l/c_s$ (sound propagation time)

mean density: $\rho_{00} = \rho_0 \frac{l_0}{l_0 + \Delta l_0}$

at $t = t_x$: $\quad l = l_0 + \Delta l_0$

***$t = t_x$:* the foils merge, weak shocks are generated**

***$t > t_x$:* expansion velocity is determined by shock hydrodynamics**

1D target expansion in planar geometry; t_x can be detected by measuring the surface velocity

An. Tauschwitz et al., NIM B, in print (2009)

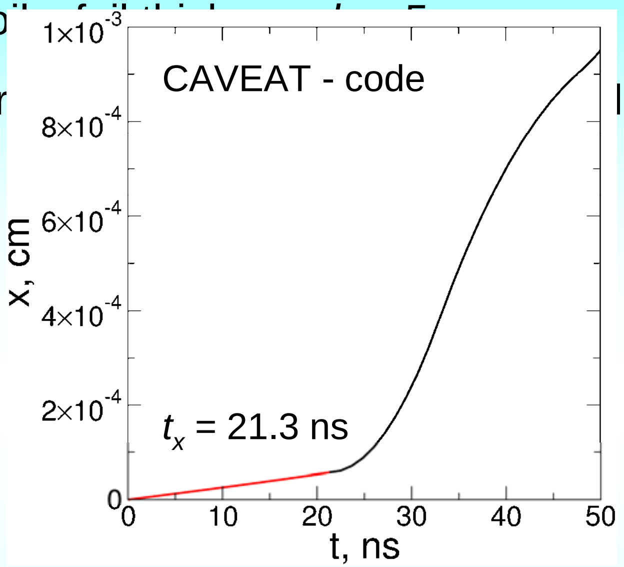
Hydrodynamic simulation

foils made of Al: ρ_0 = 2.7 g/cm^3, c_s = 5·10^5 cm/s, Γ = 2.5

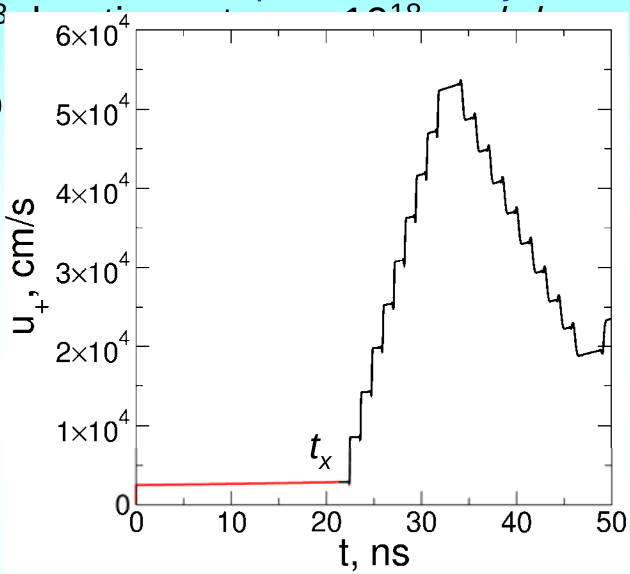
n = 10 fo … 2.2 g/cm^3 …

sound pr … l expansio

Position of the target surface



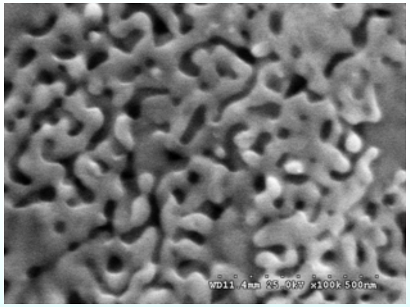
Expansion velocity

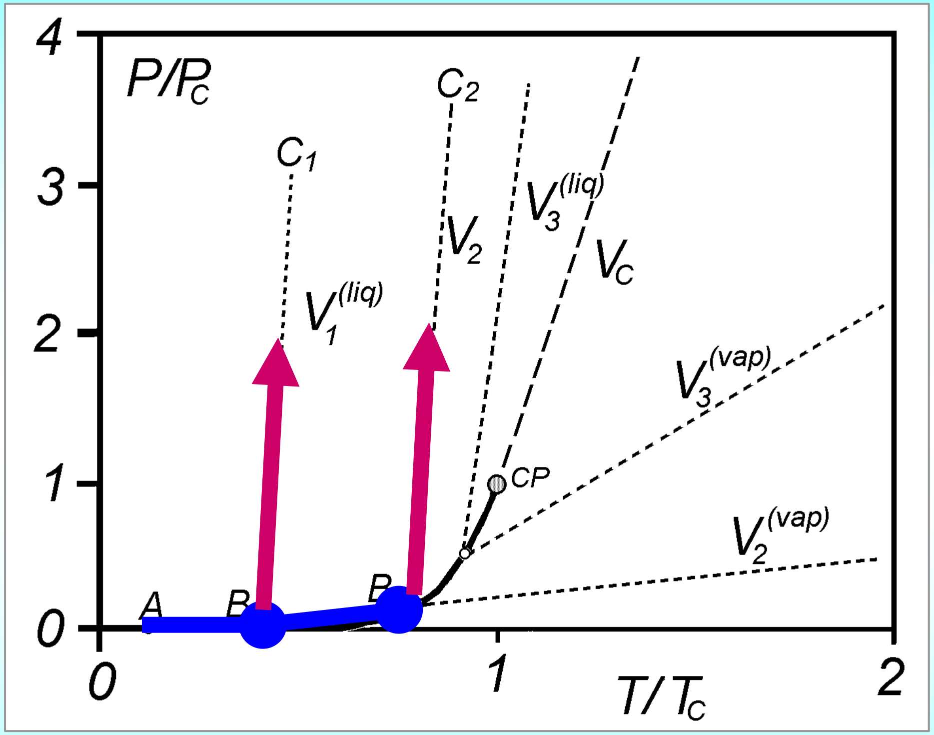


The “homogenization” time t_x can be detected by measuring the surface velocity

Anna Tauschwitz *et al.* // *Hirschegg-2009-2010*

Tracing of the Boiling Curve





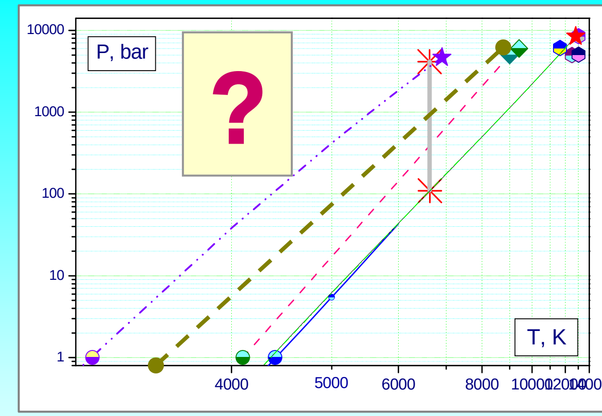
$0 < t < t*$ – **Quasi-free "isobaric" expansion**

Moment "X": $\rho = \rho*$

$t > t*$ – **Isochoric Heating + Hydrodynamic Expansion**

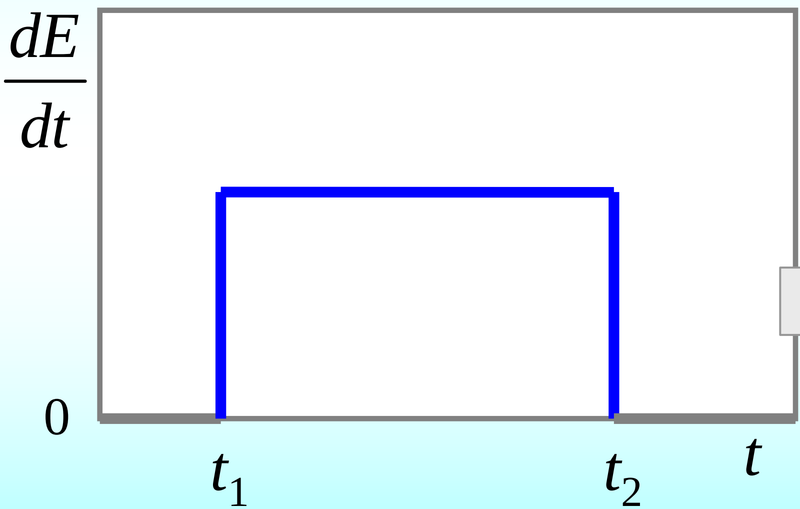
Goal for experimenters: – to catch the ***pressure jump moment*** **!**

Measurement of Uranium Vapor Pressure in Experiment with Surface Laser Heating

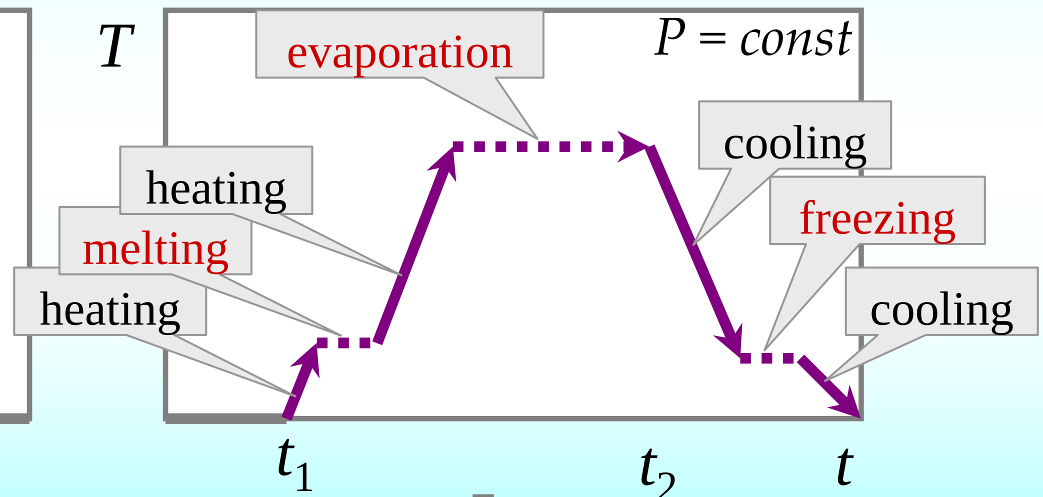


$\underline{P = const}$ *(in buffer gas)*

Laser impulse



Schematic thermogram



$P = 1 - 1000$ *bar*

$T_{melt}(P)$ $T_{boil}(P)$

Conclusions *and* Perspectives

In the case of **uranium** we meet **fundamental physical problem**:

- *Phase transition in a system with mean-particle interaction strongly dependent on density (and temperature)*

- It is **promising** to investigate this problem analytically via **simplified plasma models**

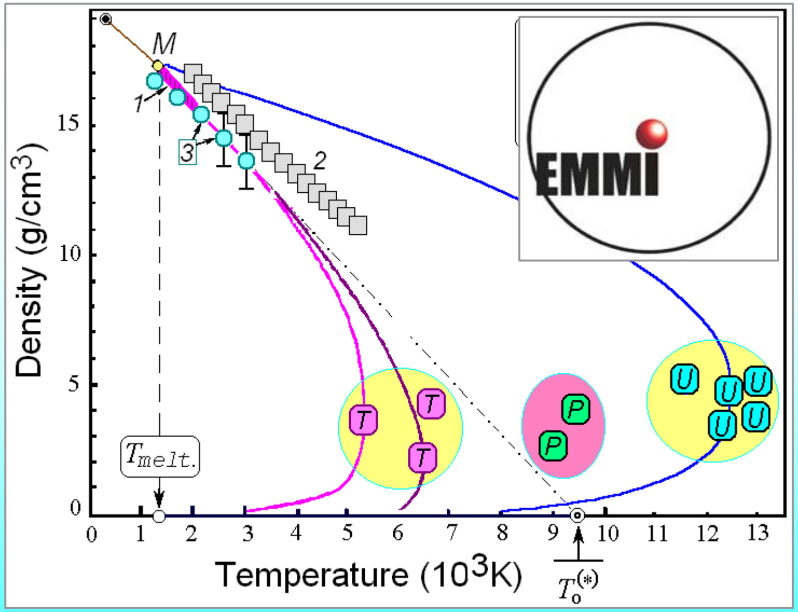
- It is **promising** to investigate this problem in **direct numerical simulation** in frames of ***ab initiio*** approaches

- It is **promising** to investigate non-congruent phase transitions **experimentally**. in particular with **intense laser** and **heavy ion** heating

! Only 1 – 3 kJ/g is needed !

Features *of* phase transitions *in* cosmic matter *and* laboratory

Thank you!



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