

X-rays as a Tool for Probing Extreme States of Matter <u>EMMI Workshop, GSI, June 2010</u>



Resolution of Uranium Critical Point Location Problem

as a goal for HIB and Laser heating and X-ray diagnostics



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Advanced nuclear reactors developments

Developments of Gas-Core Nuclear Reactor



Hypothetical "Plasma Phase Transition" in Strongly Non-Ideal Plasma



In search of "Plasma Phase Transition"

Strongly shock compressed cesium plasma



Anomalous Phase Transitions in Hydrogen and Deuterium



Norman & Starostin, Plasma Phase Transition, 1970



New Experiments (VNIIEF, Sarov)

Quasi-isentropic compressibility of deuterium <u>P ~ 100 - 300 GPa</u>



Density break in isentropic compression of gaseous deuterium ⇔ hypothetical phase transition (?)



Fortov V., Ilkaev R., Arinin V., Burtzev V., Golubev V., Iosilevskiy I., Khrustalev V., Mikhailov A., Mochalov M., Ternovoi V., Zhernokletov M. (*Phys. Rev. Lett.* 2007) // Iosilevskiy I., Gryaznov V., Fortov V. (*to be published*)

Historical comments - II

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

- Brilliant perspectives with HIB energy deposition, $\Delta E \sim 100 \text{ kJ/g.}$ but now: $\Delta E \sim 1 \text{ kJ/g}$
 - ? What could we do with $\Delta E \sim 1 \text{ kJ/g}$ and $t_{\text{HIB}} \sim 100 \text{ ns}$?

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 $\Delta E \sim 1 \text{ kJ/g}$ and $t_{\text{HIB}} \sim 100 \text{ ns}$?

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* (UO₂, UC, UN ...) *and other mixed substances*)

- applied importance
- phenomenology
- fundamental physical problem





Yu. Khariton

«Extreme State of Matter»

Int. Conferences "Equation of State" Russia, Elbrus, 1990 -2010

Int. Conferences "Khariton's Science Readings" Russian Federal Nuclear Center, Sarov, 2005 - 2010

Int. Conferences "Zababakhin's Science Readings" Russian Federal Nuclear Center, Snezhinsk, 2005 - 2010





Uranium Critical Point Location Problem

- Applied importance
- Uncertainty
- Fundamental physical problem

In cooperation with:

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

Critical Point Location Problem Uncertainty

Recommendations of IVTAN-Database (1982)

and an and a second			
Cr	9620	968	0.023
Мо	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
h	14950	438	0.072
U	11630	611	0.045
UD,	7550	122.6	0.163
UF.	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
KC1	3200	22	0.415
Rb	2106	13.22	0.246
Cs	2043	11 75	. 0.308
00	2045	11./3	01000

Critical Point Parameters

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

Critical Point Location Problem

Элемент	$T_{\rm c},{ m K}$	$p_{\rm c}, 10^8 \; \Pi {\rm a}$	$\rho_{\rm c}, \Gamma \cdot {\rm Cm}^{-3}$	$S_{\rm c}$, кал·моль ⁻¹ K ⁻¹
Zr	16250	7,52	1,79	40,45
Zn	3196	2,63	2,29	31,27
Y	10800	3,74	1,30	38,92
Yb	4280	1,38	2,36	36,92
V I	10500	10 =0		
U	11630	6,11	5,30	43,07
		,	~,~·	10,01
Ti	11790	7,63	1,31	37,92
Sn	8200	3,35	2,05	39,44
Th	14950	4,88	3,21	43,67
Tu	5910	2,65	3,22	38,52
Tl	4470	1,63	3,16	38,67
Tb	8060	3,08	2,57	42,34
Te	1850	0,75	2,21	35,22
Ta	20570	13,5	5,04	43,07
Sr	3860	0,90	0,86	35,23
Ag	7010	4,50	2,93	36,86
Se	8350	4,08	0,93	36,82
Sm	5340	2,10	2,51	40,62
Ru	15500	13,74	3,79	37,14
Rh	13510	11,23	3,62	39,66
Re	19600	15,7	6,32	43,42
Ra	3830	0,77	1,93	38,32
Pr	9160	2,85	1,86	41,39
Tc	15930	11,81	3,09	41,00
Pa	12650	4,82	3,72	43,34
Po	2050	0,62	2,67	37,52
Pt	14330	8,70	5,02	41,8
Pd	10760	7,64	3,20	36,64
Os	17110	14,49	6,83	42,60
Nb	19040	12,52	2,59	41,65
Ni	10330	9,12	2,19	36,50
Nd	7920	2,65	2,05	41,33
Mo	16140	12,63	3,18	41,21
Lu	7060	2,84	2,97	39,41
Fr	1810	0,12	0,65	39,50

Uncertainty

Fortov V., Dremin A., Leont'ev A. High. Temp. 13, (1975)

Fortov V., Khrapak A., Yakubov I. Non-Ideal Plasma Physics (2004)

Critical Point Parameters *Recommendations of IPCP RAS-GSI Database*

	Pressure	Temperatue Density		Entropy	
	P _C , kbar	Т _С , К	Q _C , g∕ñm³	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	Г
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	
Со	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	
	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

				<u> </u>		List of Uranium
7 _C ≈ 12′000 K	(t	he early estim	ation)		<u>Braut</u> (1957)	CP parameters
7 _с [К]	p _c , bar	$\rho_{\rm c} g/{\rm cm}^3$	Z _c	$ ho_{ m s}/ ho_{ m c}(*)$	References	estimations
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	- 1	-	-	-	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)	Table from:
9′400 (z _c = 3)	6000	2.59	0.706	7.32	Likalter A. (1985 - 1996)	H. Hess,
11′630	6110	5.30	0.284	3.58	Fortov V. et al. (1975)	Z. Metallkd. (2001)
11′679–12′995	-	-	-	-	Kopp I. (1967) Lang G. (1977)	Vapor Pressure and Critical Data for
12′400	4800	3.55	0.312	5.34	Morris E. (1964)	Uranium
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)	
9′636	7'700	4,50	Wide-rang	e EOS	Bushman - Lomonosov	
7′000	1'712	3,30 E	Extrapolation of	Liquid $\rho(T)$	Apfelbaum – Vorob'ev (2009)	
5'500–6'500 100 – 1'000 (<i>estimation –</i> thermal EOS calibration)) Iosilevskiy (1990)			Additions IL			
6′840 12′800	4′440 8′450	(Plasma moo (Plasma moo	del – therma del – caloric	calibratio	n) Iosilevskiy & Gryaznov SAHA-code, JNM, (2005)	



Wide-Range Equation of State

(after A. Bushman and I. Lomonosov)



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:

Semi-empirical rules:

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

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

 \Leftrightarrow

? Wrong experiment

High temperature 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 **?**



Exception

Non-congruent evaporation !

Hirschegg_2010

2009 2008

2007



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 ?



Drastic <u>*change</u> of <u><i>effective ion-ionic interaction*</u> during thermal expansion of liquid uranium and decrease of electronic degeneracy</u>

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

Uranium Critical Point Location Problem

General nature of the problem

Discussion:

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

Uranium critical point problem – is NOT exception, it is a general rule !!

Density-Temperature Diagram of Tungsten



Estimations of critical point of tungsten

- Experimental data on critical temperature [Ternovoy et al. High T-High P, V.34, (2002)] T_C ~ 13'660 ± 800 K]

Density-Temperature Diagram of Tantalum



 <u>EOS of Tantalum</u> [Fortov, Rakhel, Lomonosov et al. 1996]
 Exp – <u>experimental data</u> (exploded wire technique); *M* – melting; *D* – "diameter of two-phase boundary (wide-range EOS);
 – critical point estimations based on heat of vaporization // Alder & Young (1971) // Fortov et al.(1975) //
 – critical point estimations based on thermal expansion of liquid Ta // Young et al.(1977) // Levashov ,*Ph.D Thesis*/

Density-Temperature Diagram of Cobalt



- 1 Experimental data on density of liquid cobalt [Hess H., Kaschnitz E., Pottlacher G., (1994)];
- $T_{\rm M}$ melting temperature of cobalt (\cong 1768 K);
- T^* linear extrapolation "to-zero" of liquid density { $T^* \equiv T_M + \alpha^{-1}(T_m)$; $\alpha(T)$ thermal expansion coefficient};
- d "diameter of two-phase boundary { $d \equiv (\rho_{liq} + \rho_{gas})$ };
- 2 reconstruction of two-phase boundary and critical point estimation based on the data [1];
- 3 critical point estimation [2] based on heat of vaporization; 4 critical temperature estimation of H. Hess et
- *al.* [1] from correlation of $T_{\rm C}$ with ionization potential ("Plasma" Hypothesis /A. Likalter [3]/);

Uranium Critical Point Location Problem

In search for possible resolution of the problem:

(A) Wrong experiments (!?)

- Shaner et al.-1977 // Shpilrain et al. -1987 // Mulford & Sheldon -1991 // Boiveneau et al.-1993....

- (B) Anomalous (*non-convex*) form of density-temperature coexistence curve
- (C) Anomalous large <u>upward non-linearity</u> of saturation curve in $(\ln p_S 1/T_S)$ coordinates
- (D) Anomalous <u>extra-low value</u> of the <u>critical compressibility factor</u> of uranium, $(pV/RT)_{c} \ll 1$.
- (E) Anomalous <u>extra-low value</u> of the uranium <u>ratio</u> of <u>critical to normal</u> densities, $(\rho_{\rm C} / \rho_0) \ll 1^{-1}$

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

Uranium Critical Point Location Problem

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»

More exotic and hypothetical

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 UO₂ \Leftrightarrow **the same physical problem**

Phase transition in the system with varying composition



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 <u>*uniformly-compressible*</u> 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
 <u>without</u> intermediate <u>hydrodynamic</u> <u>re-calculations</u>
- 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

In this moment we obtain:

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





It means :

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

Pressure Jump

Moment "X"

* * * * * * * * * * *

Pressure Jump

If we catch this moment

- and if we know:
- 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.



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

Quasi-static heating of a stack target



 $t > t_x$: 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



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

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

Thermophysical investigations via HIB (novel regimes)

Tracing of the Boiling Curve



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





Measurement of Uranium Vapor Pressure in Experiment with Surface Laser Heating

dE

dt

0



10000 -



EMMI: Cosmic Matter in the Laboratory **Plasma Physics** *with* **Intense Laser** *and* **Heavy Ion Beams** <u>*3rd Workshop: Moscow, May 20-21, 2010*</u>



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







<u>Support</u>: RAS Scientific Program "Physics of Extreme State of Matter" MIPT Research-Education Center "Physics of Extreme State of Matter" ExtreMe Matter Institute - EMMI

Выпуклость фазовой границы $\rho(T)$

(*правило «Бермудского» треугольника*)

- **Плотность** конденсированного состояния (кристалла и жидкости), как функция температуры, **<u>линейна</u>** на большей части границы сублимации (*m*в) и кипения (*ж*)

ρ(*T*)-граница двухфазной области всегда выпукла (эмпирическая закономерность)

Эта граница всегда лежит внутри треугольника (эмпирическая закономерность)



Hypothetical "Plasma Phase Transition"

in Strongly Non-Ideal Ionic Plasma



Anomalous Phase Transitions in Hydrogen and Deuterium

1968 - 1970

Норман и Старостин, Плазменный Фазовый Переход

1972 С.Б. Кормер с сотр. (ВНИИЭФ, Саров), Аномальный скачок плотности при изоэнтропическом сжатии водорода до давлений P~ 3 Mbar

? - Plasma Phase Transition - ?

