



### Non-Congruent Phase Transitions in Cosmic Matter and Laboratory



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## Non-Congruent phase transition – – what does it mean ?

Non-congruence – phase coexistence with different chemical composition !

**Evident definition – in terrestrial applications** 

**Non-evident – in interiors of compact stars** 

Non-evident – in ultra-high energy ion collisions products

#### <u>The base</u>

#### Non-Congruent Phase Transition in Uranium Dioxide

Hypothetical severe accident at fast-breeder nuclear reactor

<u>Support</u> Vladimir Fortov (*Russia*) Claudio Ronchi (*Germany*) Boris Sharkov (*Russia*) Dieter Hoffmann (*Germany*)

INTAS 93-66 // ISTC 2107 // CRDF MO-011

<u>Research Programs of</u> <u>Russian Academy of Science:</u>

"Physics and Chemistry of Extreme States of Matter" and "Physics of Compressed Matter and Interiors of Planets" <u>Cooperation</u>

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### Non-Congruent Phase Transition in Uranium Dioxide



#### **INTAS Project** (1995–2002)

<u>Cooperation</u>: MIPT – IHED RAS – IPCP RAS – OSEU – MPEI ⇔ ITU (JRC, Germany)

Project Coordinator – C. Ronchi (ITU, JRC) ⇔ Project Supervisor – V. Fortov

#### **ISTC Project** (2002–2005)

GSI

Institute for Transuranium Elements

IHE

<u>Cooperation</u>: MIPT – IHED RAS – IPCP RAS – ITEP – VNIIEF ⇔ GSI (JRC, Germany)

Project Manager – B. Sharkov (ITEP, Moscow) ⇔ Project Science Supervisor – V. Fortov

ITEP (Moscow)

## **Two problems in phase transition calculation**

# Construction of Equation of State (EOS) Phase coexistence parameters calculation

#### **Chosen approach and fundamentals**

## **Sketch of theoretical approach**



\* Iosilevski I., Yakub E., Hyland G., Ronchi C. Trans. Amer. Nuclear Soc. 81, 122 (1999)

\* Iosilevski I., Yakub E., Hyland G., Ronchi C. Int. Journal of Thermophysics 22 1253 (2001)

\* Iosilevskiy I., Gryaznov V., Yakub E., Ronchi C., Fortov V. Contrib. Plasma Phys. 43, (2003)

\* Ronchi C., Iosilevskiy I., Yakub E. Equation of State of Uranium Dioxide / Springer, Berlin, (2004)

\* Iosilevskiy I., Son E., Fortov V. Thermophysics of non-ideal plasmas. MIPT (2000); FIZMATLIT, (2009)

### **Quasi-chemical representation** ("Chemical picture")



## Phase coexistence parameters calculation

(two approaches)

Ordinary way:Maxwell ("equal squares") construction{in unique two-phase pressure-density:  $P(V)_T$ }Or"Double tangent" construction{in free energies of two phases:  $F_1(V) \Leftrightarrow F_2(V)$ }



**Congruent evaporation in U-O system** <u>does not correspond to the total equilibrium</u> (only to the partial one)

# <u>Maxwell approach</u> - should be rejected as non-adequate

## *Correct approach*:

- Gibbs (+ Guggenheim) conditions

#### Phase equilibrium conditions in reacting Coulomb system



(see for example: Iosilevskiy I., Encyclopedia on low-T plasmas. III-1 (suppl) 2004, P.349-428)

#### **Electrostatics of phase boundaries in Coulomb systems**



#### **Electrostatics of Quark-Hadron Interface**



Nuclear Crust on Strange Mater



After Fridolin Weber, WEH Seminar, Bad Honnef, 2006

#### **Gibbs - Guggenheim conditions in reacting Coulomb system**

#### **Non-congruent evaporation in U-O system** (*Gibbs - Guggenheim conditions*)



## End-Points of Non-Congruent Phase Transition



## <u>NB</u> !

- Point of temperature maximum
- Point of pressure maximum
- Critical point

are three different points !

## Non-congruent phase transformation in two-phase region



Oxygen depleted liquid ! *Different stoichiometry*!



#### Last vapor bubbles in boiling liquid



Oxygen enriched vapor ! Different stoichiometry! Non-congruent evaporation in U – O system

## **Isotherms in two-phase region**



• Isothermal phase transition starts and finishes at *different pressures* 

• Isobaric phase transition starts and finishes at *different temperatures* 

## Chemical composition at coexisting phases



Liquid  $(O/U = 2.0) \Leftrightarrow$  Vapor (O/U > 2.0)

Vapor (O/U = 2.0)  $\Leftrightarrow$  Liquid (O/U < 2.0)

Non-congruent evaporation in U – O system

### Isobaric transition through the two-phase region



## EMMI : Cosmic Matter in the Laboratory

## Non-congruence in general

Main issue for study of non-congruent evaporation in U–O system

## Non-congruence of phase transition in U-O system – – is it an exception or a general rule ?



Non-congruence in H<sub>2</sub>O etc... – what does it mean **?** 



<u>BASIC STATEMENT</u>: Any phase transition in a system of two or more chemical elements must be <u>non-congruent</u>



### Neptune and "hot-water" extrasolar planet GJ436b



Any phase transition in *high-T\_high-P* water must be *non-congruent* 

#### Plasma Phase Transitions in H<sub>2</sub> + He plasma

(planetary science)



Contrib. Plasma Phys. 35 (1995) 2, 109-125

Plasma Phase Transition in Fluid Hydrogen-Helium Mixtures

M. SCHLANGES (a), M. BONITZ (b), and A. TSCHTTSCHJAN (b)





### Plasma Phase Transitions in H<sub>2</sub> + He plasma



Fig. 7. Coexistence pressure for H-He mixtures for different values of the mixing parameter, for the hydrogen-like plasma phase transition and for the helium-like plasma phase transition.

### Thermodynamics of H<sub>2</sub> + He plasma (planetary science)

0



**Congruent** phase transition in the H<sub>2</sub>+ He plasma

**Non-Congruent** phase transition in the H<sub>2</sub>+ He plasma



#### Thermodynamics of H<sub>2</sub> + He plasma (planetary science)

0



M. SCHLANGES (a), M. BONITZ (b), and A. TSCHTTSCHJAN (b)



Fig. 7. Coexistence pressure for H-He mixtures for different values of the mixing parameter, for the hydrogen-like plasma phase transition and for the helium-like plasma phase transition.

Phase diagram in simple mixture H<sub>2</sub> + He could be complicated due to non-congruence

**The question is:** 

What kind of phase transition one can expect in high-*T* high-*P* complex plasma ?  $H_2 + He + H_2O + NH_3 + CH_4...$ at *T* ~ 1 – 20 kK & *P* ~ 1 – 10 Mbar

#### **Typical composition in planetary science**

### Hypothetical <u>non-congruent</u> plasma phase transition in H<sub>2</sub> + He mixture in interiors of Jupiter and Saturn



**The question is:** 

## What kind of phase transition one can expect in high-*T* high-*P* complex plasma ? $H_2 + He + H_2O + NH_3 + CH_4...$ at *T* ~ 1 – 20 kK & *P* ~ 1 – 10 Mbar

#### We know almost nothing about it, but we should know everything . . . .

# Cassini-Huygens

#### **Conclusions** and **perspectives**

- Non-congruence of phase transitions in H<sub>2</sub> / He mixture can 'provoke' to the H⇔He separation in interiors of <u>Jovian</u> and <u>Extrasolar</u> planets and <u>Brown Dwarfs</u>.
- New experiments are desirable for study of discussed <u>non-congruence</u> for phase transition in H<sub>2</sub> / He / H<sub>2</sub>O / NH<sub>3</sub> / CH<sub>4</sub> / mixture.



# Hypothetical non-congruent phase transitions (*short list*)

#### Terrestrial applications:

- Uranium- and Plutonium-bearing compounds:
  - UO<sub>2</sub>, PuO<sub>2</sub>, UC, UN, ... ets.,
- Metallic alloys: (Li-K-Na,...etc.)
- **Oxides**: (SiO<sub>2</sub>...etc.)
- Hydrides of metals (LiH,... etc.)
- Ionic liquids and molten salts:
   alkali halides (NaCl, ... etc.), ammonium halides (NH<sub>4</sub>Cl ... etc.)
  - "Dusty" and Colloid plasmas:
     (Coulomb system of macro-ions + Z and micro-ions: +1, -1)

Non-Congruence in Cosmic Matter:

- Plasma Phase Transitions in mixture: H<sub>2</sub>/ He /H<sub>2</sub>0 / NH<sub>3</sub> / CH<sub>4</sub> in Giant Planets, Brown Dwarfs and Extra-Solar Planets,
- Phase Transitions in White Dwarfs,
- Phase Transitions in Neutron Stars,
- Phase Transitions in "Strange" Stars (quark-hadron transition ... ets.)

Iosilevskiy I. / Int. Congress on Plasma Physics / Fukuoka, Japan, 2008 (J. of Plasma and Fusion Research, 2009)



# Non-congruence in exotic situations

# Non-congruence in compact stars

**The New Physics of Compact Stars** 



## <u>Compact stars</u>

White dwarfs, Neutron stars, "Strange" (quark) stars, Hybrid stars



Рис. 65. Массы планет (в единицах массы Земли) и их среднее расстояние от Солнца [371]

Hybrid ("strange") white dwarfs

Mathews G., Weber F. et al. J. Phys. G, 32, (2006) - White dwarfs with strange-matter cores



#### Hypothetical phase transitions in interior of compact stars: are they <u>CONGRUENT</u> or <u>NON-CONGRUENT</u> ?







#### First quark droplets in hadron matter





Last hadron bubbles in quark matter

## Structured Mixed Phase Concept $\Leftrightarrow$ "Pasta"



Schematic picture of pasta structures. Phase transition from blue phase (left-bottom) to red phase (right-bottom) is considered.

Pasta structures in compact stars /arXiv:nucl-th/0605075v2 /2006/

Maruyama T., Tatsumi T., Endo T., Chiba S.



## Structured Mixed Phase $\Leftrightarrow$ "Pasta" plasma

**'Pasta' plasma** – hadron-quark phase transition <u>in interior of neutron stars</u> ('Mixed phase' of Glendenning *et al.* 1992)

- Charged quark droplets (rods, slabs) in equilibrium hadron matter
- Charged hadron bubbles (tubes, slabs) in equilibrium quark matter



Fig. 1. Nuclear and quark matter tructures in a  $\sim 1.4 M_{\odot}$  neutron star. Typical sizes of structures are  $\sim 10^{-14} m$  but have been scaled up to be seen.

Heiselberg and Hjorth-Jensen <u>Phase Transitions in Neutron Stars</u> arXiv:astro-ph/9802028v1 (1998)

T.Maruyama, T.Tatsumi, T.Endo, S.Chiba <u>Pasta structures in compact stars</u>

arXiv:nucl-th/0605075v2 31 (2006)

"Pasta" plasma:- "Spaghetti" phase, "Lasagne" phase . . . .

Non-congruence in exotic situations (di scussi on)



# **Quark-hadron** phase transition via "mixed-phase" scenario has the features of non-congruent PT !





# Hypothetical phase transitions in ultra-dense matter:



#### Evaporation of strange lumps in the early Universe *Alcock C., Farhi E.* (*PRD, 1985*) *Alcock C., Olinto A.* (*PRD, 1989*)

Strange matter, a stable form of quark matter containing a large fraction of strange quarks, may have been copiously produced when the Universe had a temperature of  $\sim 100$  MeV. We study the evaporation of lumps of strange matter as the Universe cooled to 1 MeV. Only lumps with baryon number larger than  $\sim 10^{-2}$  could survive. This places a severe restriction of scenarios for strange-

Strange matter is a form of quark matter that has been conjectured to be stable at zero temperature. If heated to a temperature  $T \ge 2$  MeV, a strange-matter lump evaporates nucleons from its surface. We show that at higher temperatures  $T \ge 20$  MeV, strange matter boils, with bubbles of hadronic gas forming and growing throughout the interior. Strange matter, or any other phase which resembles strange matter, could not have survived this process in the early Universe.





#### Hypothetical phase transitions in ultra-dense matter

are they CONGRUENT or NON-CONGRUENT ?



# The problem of non-congruence for the Quark-Hadron phase transition is relevant !

Iosilevskiy I. / Int. Workshop "Physics of HEDM", JINR, Dubna, Russia, 2008

# Non-congruence in high-energy collisions



After Markus H. Thoma, SCCS, Moscow, 2005

## EMMI : Cosmic Matter in the Laboratory

Hydrodynamics of expanding fireball when it crosses quark-hadron phase boundary depends significantly from the fact – is this phase transition congruent or non-congruent ! EMMI : Cosmic Matter in the Laboratory

#### **The question is:**

## What kind of phase transition one can expect in high-*T*\_high-*P* complex plasma ? SiO<sub>2</sub> + FeO + Al<sub>2</sub>O<sub>3</sub> + CaO + . . .



#### **Exploration of the Moon Continues!**



LCROSS Lunar CRater Observation and Sensing Satellite

What kind of phase transition one can expect in high- $T_high-P$ complex plasma? SiO<sub>2</sub> + FeO + Al<sub>2</sub>O<sub>3</sub> + CaO  $T \sim eV \& P \sim GPa$ 

The question is open

#### <u>NB</u> !

Phase transition in each constituent (SiO<sub>2</sub>, FeO, Al<sub>2</sub>O<sub>3</sub>, CaO...) must be *non-congruent* !

Phase transitions in the mixture <sup>11</sup> must be *non-congruent* moreover !



#### Features of isentropic release crossover of non-congruent phase transition boundary

ISTC: UO<sub>2</sub> isochoric heating under heavy ion beam irradiation



"Retrograde regime" – typical scenario for transition through the two-phase region of **non-congruent** phase transformation

## EMMI : Cosmic Matter in the Laboratory

#### **Conclusions** and **Perspectives**

- Non-congruent phase transition is general phenomenon.

- Non-congruent phase transition is universal phenomenon.

- Non-congruent phase transition is interesting phenomenon.

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

- It is **promising** to investigate non-congruent phase transitions in **direct numerical simulations** ("numerical experiment") DFT\_MD, PIMC, WP\_MD...

- If one takes into account hypothetical **non-congruence** of **phase transitions** in **cosmic matter** objects (*planets, compact stars etc.*) he should **revise** totally the **scenario** of all **phase transformations** in these objects.



Non-Congruent Phase Transitions in Cosmic Matter and Laboratory

# Thank you!



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"Physics and Chemistry of Extreme States of Matter" and "Physics of Compressed Matter and Interiors of Planets"



## Hypothetical phase transition in H<sub>2</sub>/He mixture

Giant planets evolution problem

after Chabrier G., Saumon D., Hubbard W., Lunine J. (SCCS-1992, Rochester)

Cassini-Huygens



Fig. 1. Pressure and density profiles of optimized models of Jupiter (top panel) and Saturn (bottom panel), plotted as a function of mean radius. Discontinuities in the density clearly mark the boundaries of the four layers of the models: rocky core, ice manule, metallic and molecular



#### Hypothetical phase transitions in interiors of GP-s and BD-s via "additivity approximation"





Presence of helium relax phase transition in hydrogen <> presence of hydrogen relax phase transition in helium

Schlanges M., Bonitz M, Tschetschjan A. Contrib. Plasma Phys. 35 109 (1995)

(\*) Pfaffenzeller O. et al. PRL 74 (13) 2599 (1995)



## Giant planets interior composition





## Five layers (!) model of Saturn's interior

Model $Y_0$ $Y_2$ $Z_{2-4}$ $P_{1-2}$ $M_{He}$ , core $M_{core}$	
$Y_1 = 0.06, Y_2 = 0.25, Z_1 = 0.02, I/R = 2.2$	
MS1 0.267 0.00 0.30 3.0 0.42 10.66 16.18	1 bar 135 K
MS2 0.171 0.00 0.40 3.0 0.64 4.58 8.59	1.601, 10011
MS3 0.225 0.00 0.30 2.0 0.44 9.88 15.06	
MS4 0.133 0.00 0.40 $\blacksquare 2.0$ 0.67 4.02 7.65	
MS5 0.274 0.25 0.30 3.0 0.46 6.33 9.99	
MS6 0.187 0.25 0.40 3.0 0.72 0.05 1.03	
MS7 0.285 0.25 0.25 2.0 0.43 7.34 10.74	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	/lbar, <b>4′000</b> K
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	5′500 K
$\frac{300}{100} = \frac{300}{100} = $	
MS15 $0.249$ H <sub>2</sub> O 5 $0.007$ $0.76$ (b)	
$Y_3 = 0.25$	
$Y_1 = 0.10, Y_2 = N \Pi_3 2.2$	
MS16 0.275 CH 3 10.91 16.54	
MS17 0.186 4 3 4.98 9.25 0.23 24 2.39 8 8 Mbar 7/600 k	C 1
MS18 0.234 2 10.22 15.55 0.0 Mbar, 7000 0.0 Mbar, 7	
MS19 0.149 (Fe + NI) 7 4.50 8.45 $e=2$ : IR+He - 4 3(6.03) 1 (16 Mber 0/000 K	
MS20 0.282 3 6.69 10.51 (0.09) 8.07 (8.02) (12. noar, synu) 10 MiDal, 9'000 K	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	
MS22 0.277 00.21 0.27 2.0 0.56 6.59 9.7 18 Mbar, 9'400 K	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	
MS25 0.263 0.25 0.25 1.5 0.66 5.45 8.41	
$MS26  0.327  0.35  0.25  3.0  0.43  7.93 \qquad 11.52$	N.1.1
$MS27  0.287  0.31  0.30  3.0  0.56  4.16  6.89  I = "ICeS" (H_2O, NH_3, CH_4)  R = ROCKS + Fe$	+ INI
MS28 0.248 0.35 0.35 3.0 0.71 0.90 2.34	
MS29 0.301 0.35 0.25 2.0 0.57 4.39 6.80 $Y_{\rm e} = \text{mass fraction He}$	
MS30 0.266 0.35 0.30 2.0 0.73 1.36 2.89	
MS31 0.291 0.35 0.25 1.5 0.71 3.13 5.12 7 - mass fraction (HONH CH - Fo	
MS32 0.259 0.35 0.30 1.5 0.87 0.48 1.64 $Z_{i} = 1135511301011(1_{2}O, 1NT_{3}, CT_{4} + PC)$	T NI)



Non-congruent phase transitions in astrophysical objects



# H<sub>2</sub>O, CH<sub>4</sub>, NH<sub>3</sub> in giant planets



After N.Nettelmann, R.Redmer et al. (2007)

Chemical composition of Neptune [1]:

- 56% water
- 36% methane
- -8% ammonia

W. Hubbard. Science, 214 (1981)

#### "Hot-water" extrasolar planet GJ436b