ЗАБАБАХИНСКИЕ НАУЧНЫЕ ЧТЕНИЯ 2019

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Atomistic simulations of defects formation at swift heavy ion irradiation in U-Mo alloy

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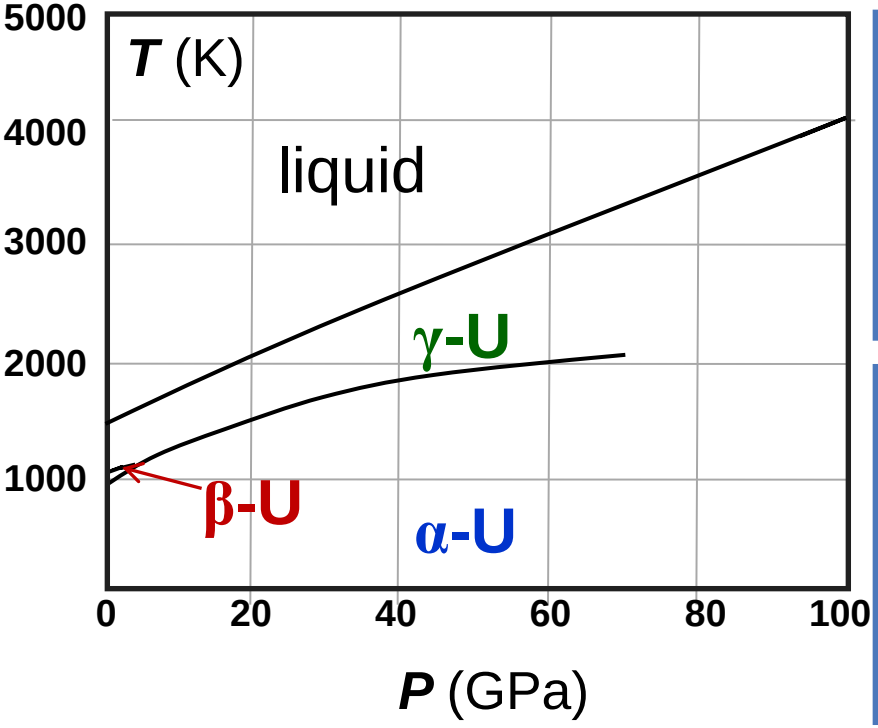
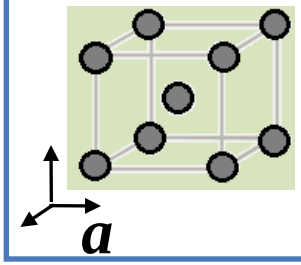
MIPT JIHT

Contents

- Motivation
- U-Mo alloy structures
- U-Mo alloy temperature-concentration diagram
- Defect formation at heating/melting of ionic subsystem
- The threshold stopping power of swift ions dependence on temperature in γ-phase

Phase diagram of pure uranium





γ-U **Body centered cubic**

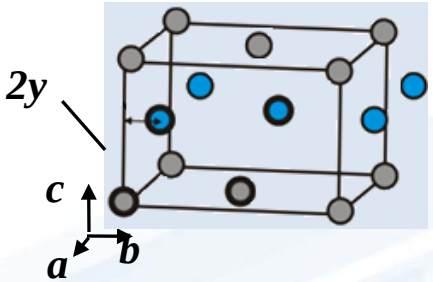
with 2 atoms in the unit cell

T = 1045 K – 1408 K

α-U **Orthorhombic**

4 atoms in the unit cell

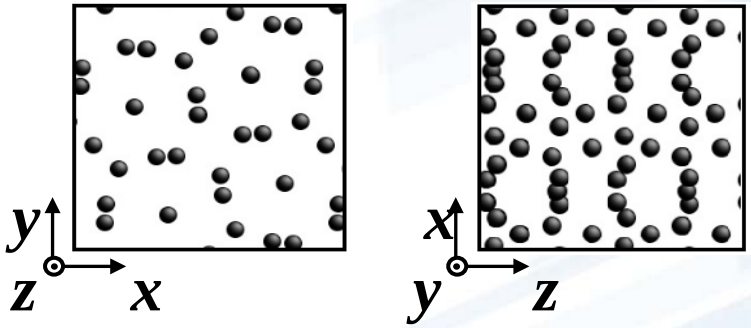
T = 300 K – 945 K



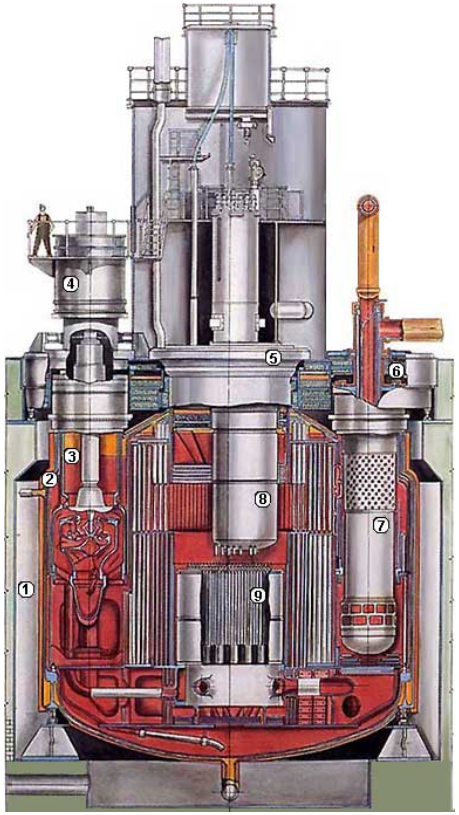
β-U **Tetragonal**

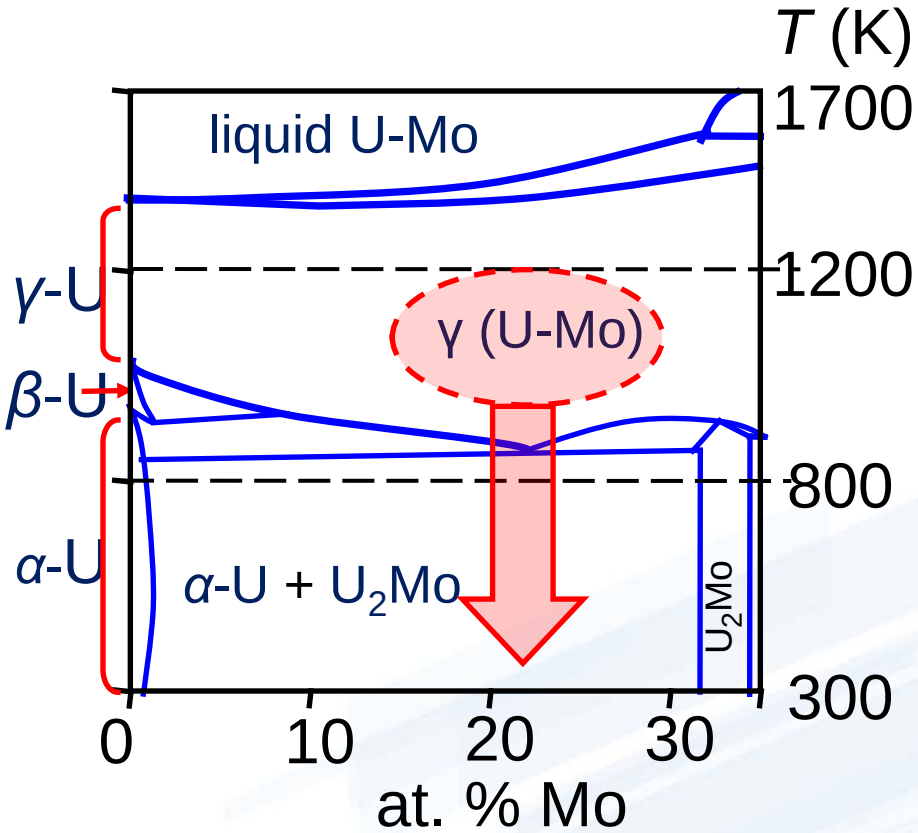
with 30 atoms in the unit cell

T = 945 K – 1045 K, *P* < 3 GPa



U-Mo alloys



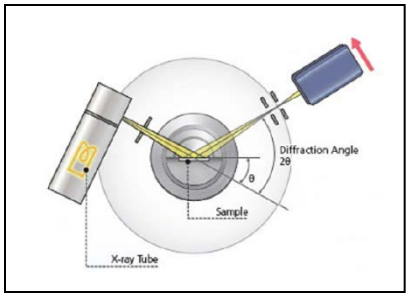


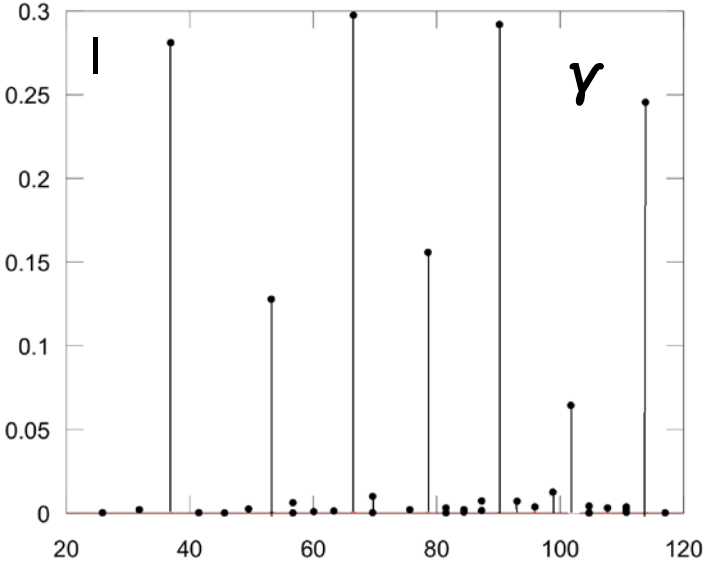
Uranium metal alloys are the most prominent candidates for advanced fast-neutron reactors (Gen IV)

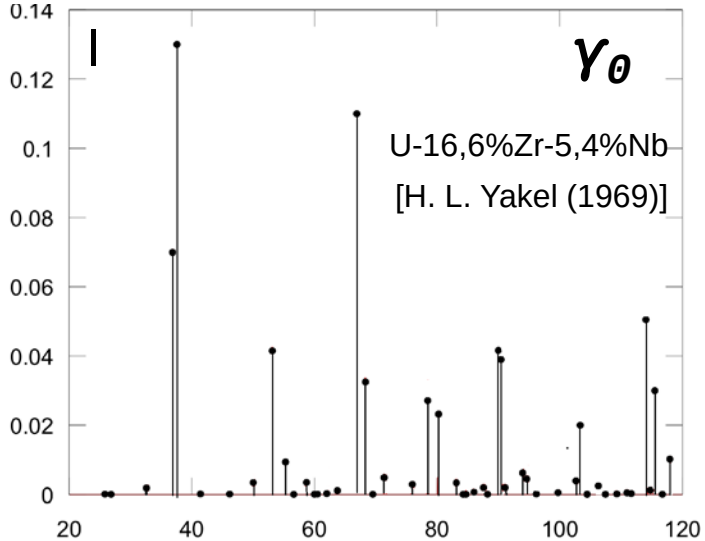
[1] *Kim Y* Comprehensive nuclear materials, volume 3: Advanced Fuels/Fuel Cladding/Nuclear Fuel Performance Modeling and Simulation (Elsevier Ltd.) 2012

[2] *Sinha V*, *Hegde P*, *Prasad G*, *Dey G* and *Kamath H* Journal of Alloys and Compounds 506 253–262 2010

Anisotropy of the U-Mo alloy







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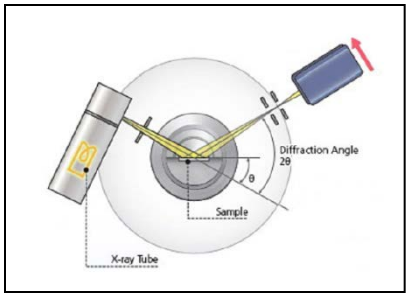
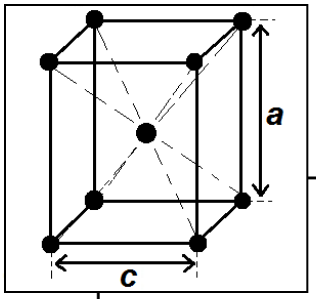
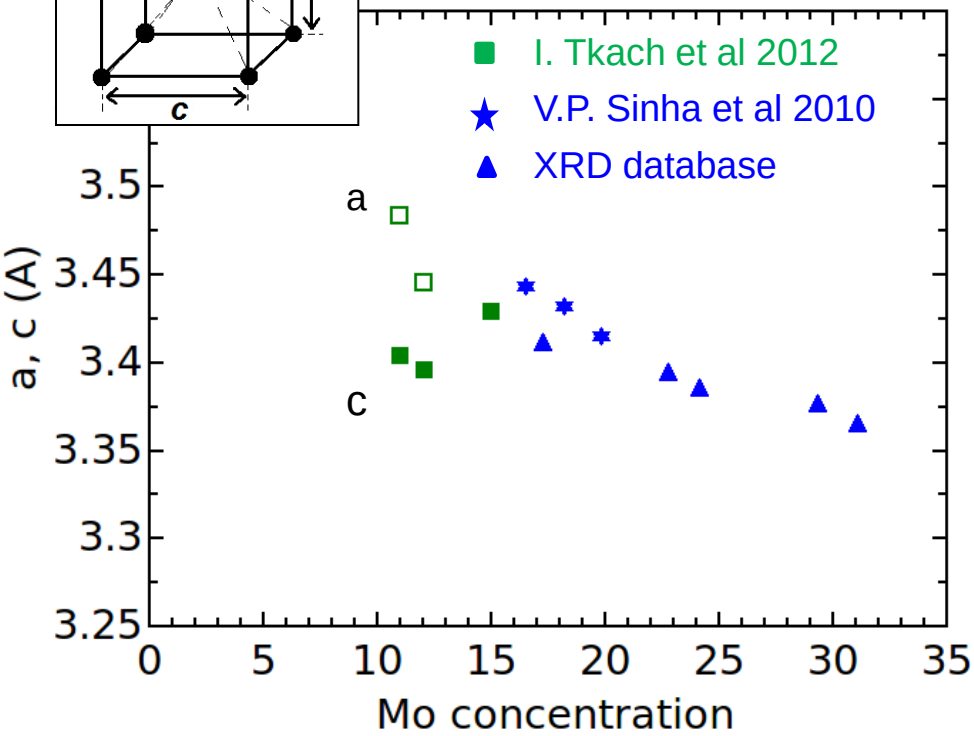
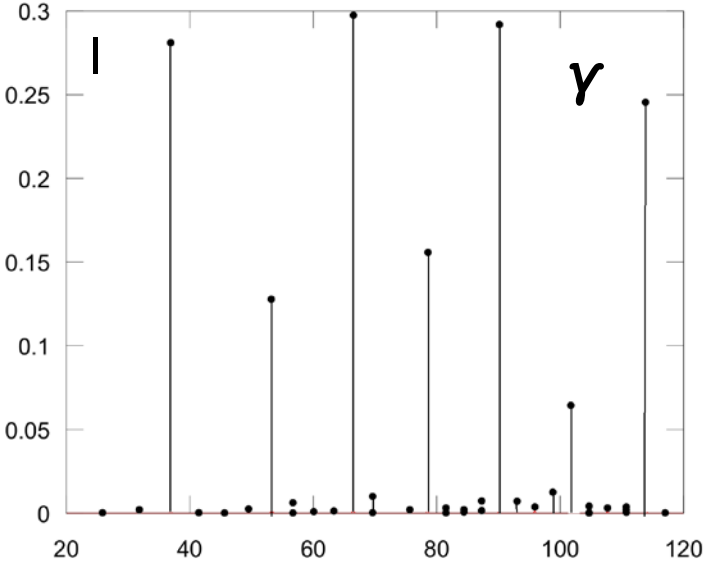
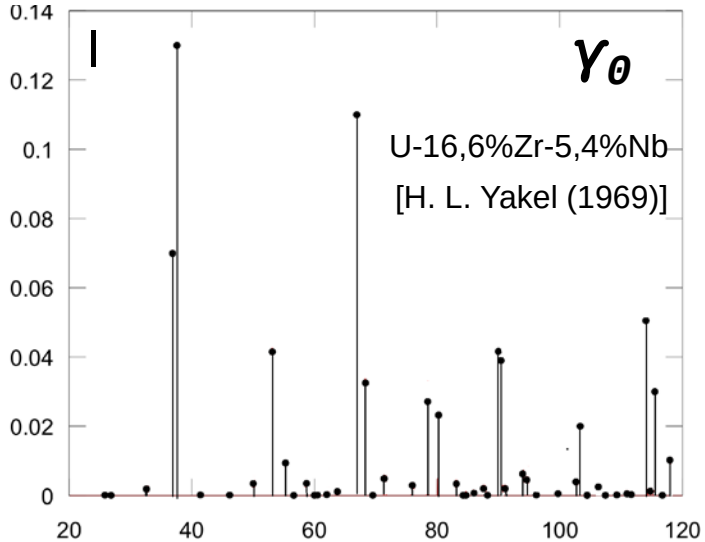






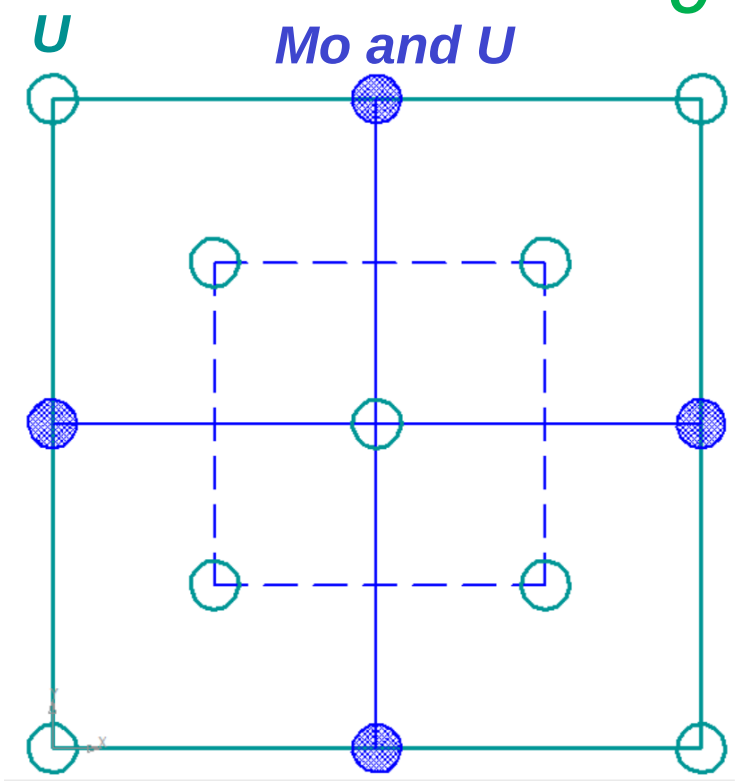




U-Mo alloy structures

γ^0 structures

$c < a = b$

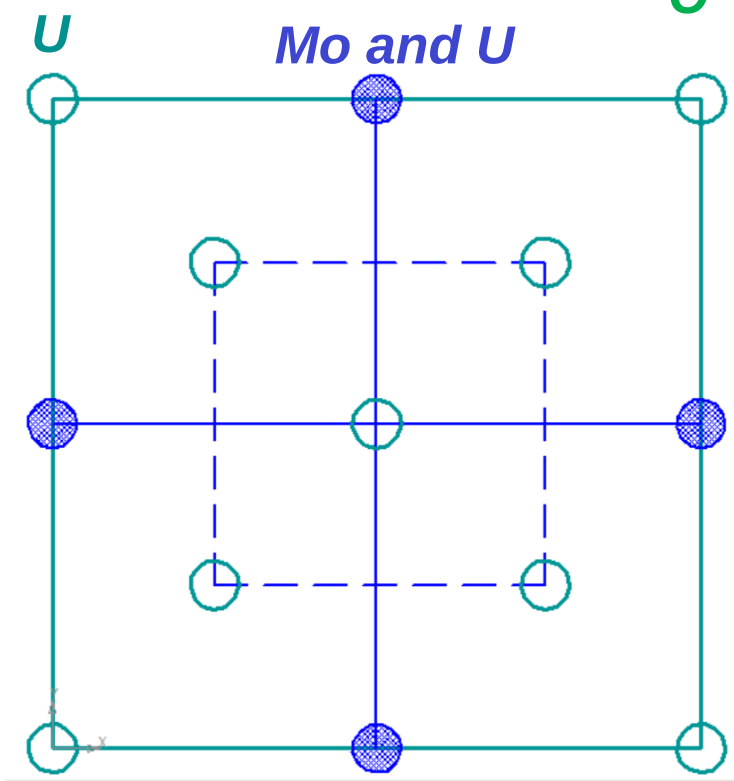


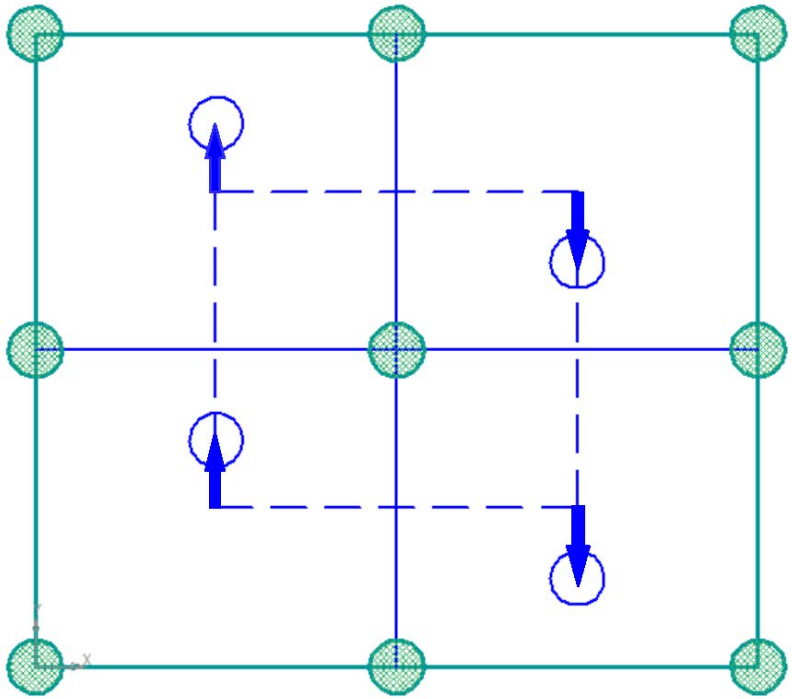
Mo and U atoms ordering

K. Tangri (1961)
B.W. Howlett (1970)

γ^0 structures

$c < a = b$





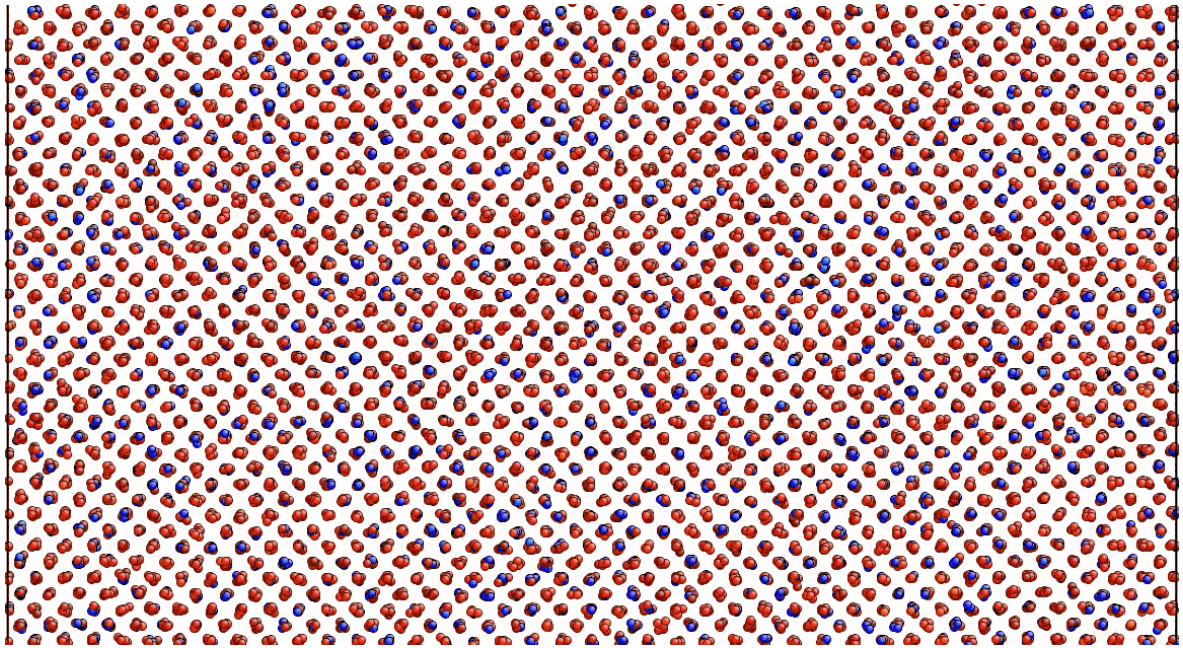
Mo and U atoms ordering

K. Tangri (1961)
B.W. Howlett (1970)

Central atom displacement

H.L. Yakel (1969)
B.W. Howlett (1970)

Preparing the equilibrium γ^0 U-Mo structure



BCC lattice

a_0 = 3.45Å

Mo – blue atoms

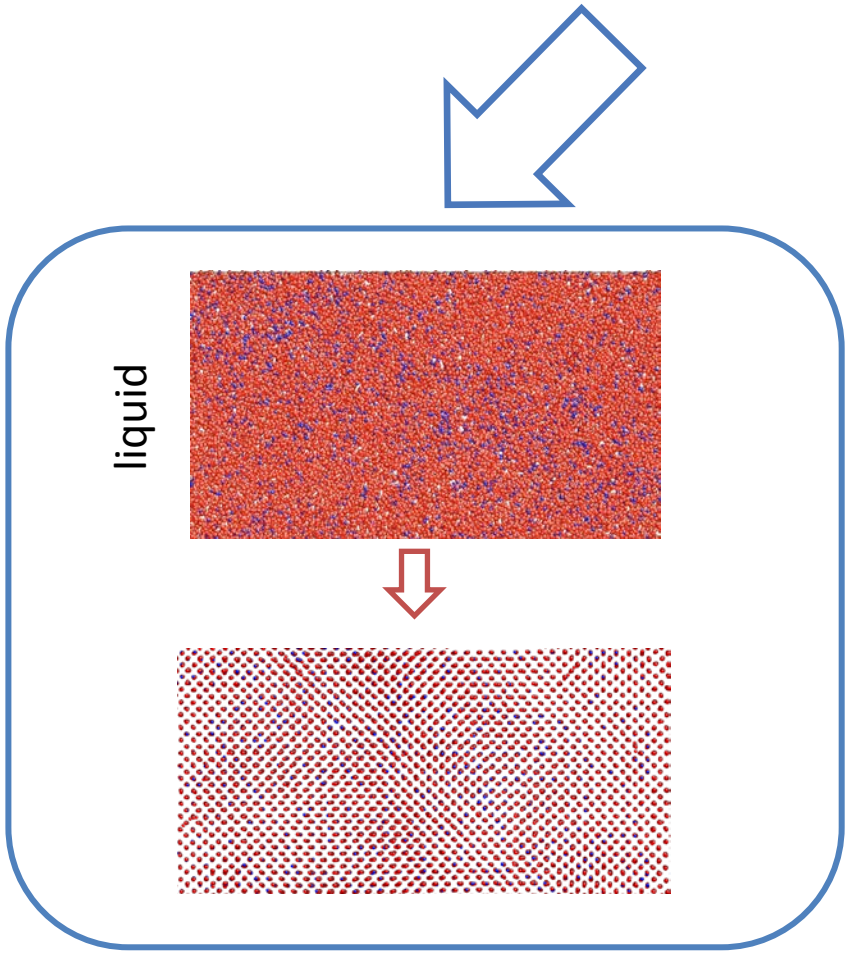
U – red atoms

ADP potential

[Starikov et al JNM 2018. V. 499. P. 451.]

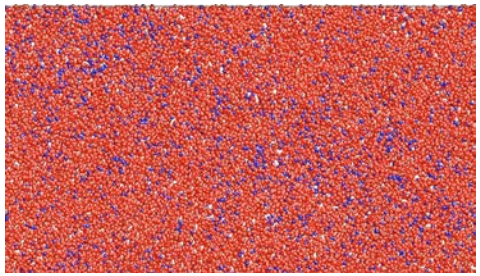
$$E_{tot} = \frac{1}{2}\sum_{i,j(j\neq i)} \Phi_{ij}(r_{ij}) + \sum_i F_i(\bar{\rho}_i) + \frac{1}{2}\sum_{i,\alpha}(\mu_i^\alpha)^2 + \frac{1}{2}\sum_{i,\alpha,\beta}(\lambda_i^{\alpha\beta})^2 - \frac{1}{6}\sum_i v_i^2$$

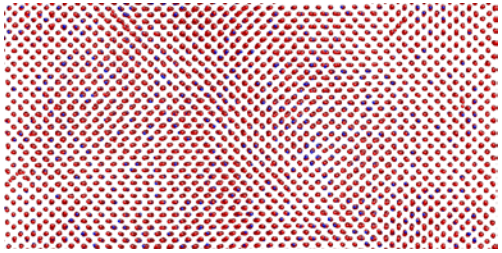
Preparing the equilibrium γ^0 U-Mo structure

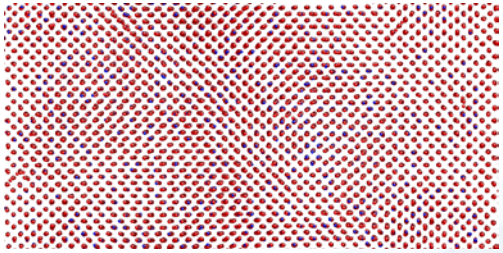


Preparing the equilibrium γ^0 U-Mo structure

liquid







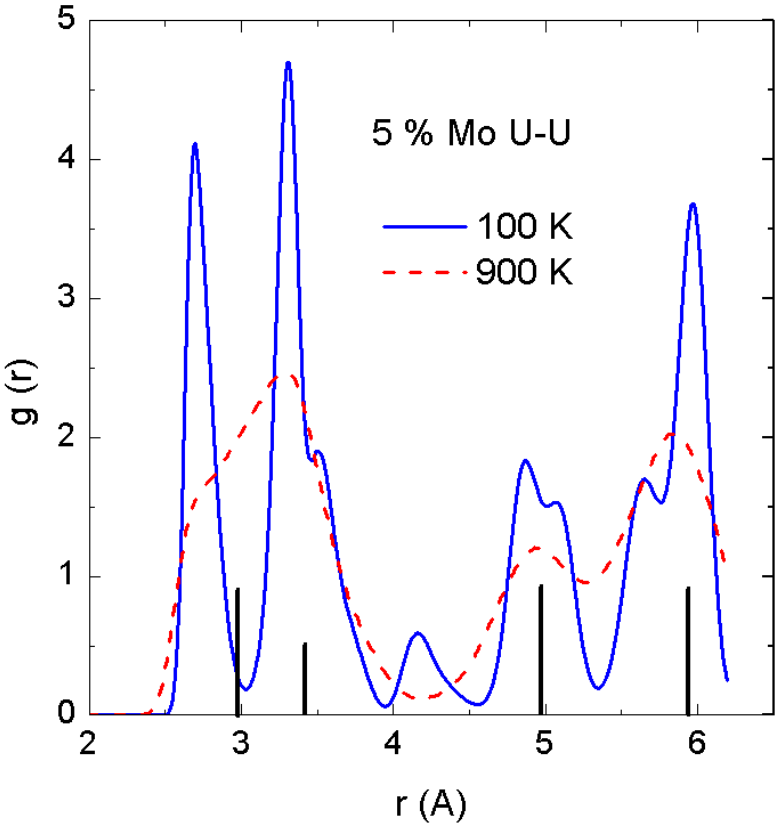
Random distribution of Mo atoms

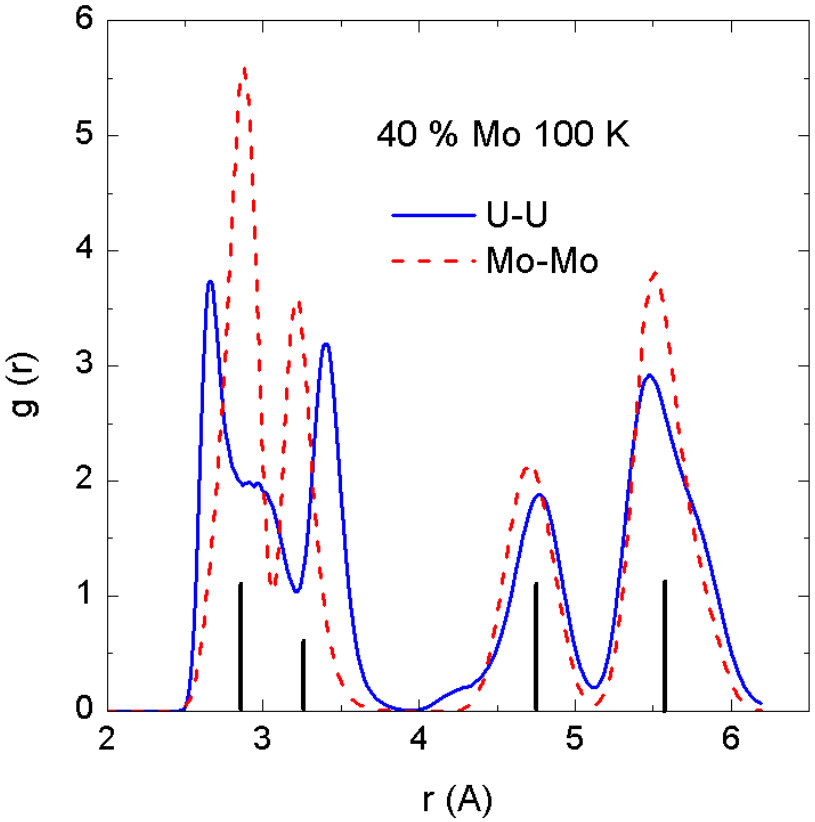
P = 0

There is no long-range ordering by the type of atoms in crystal lattice. The lattice parameters and the local structure of the alloy are independent of short range order

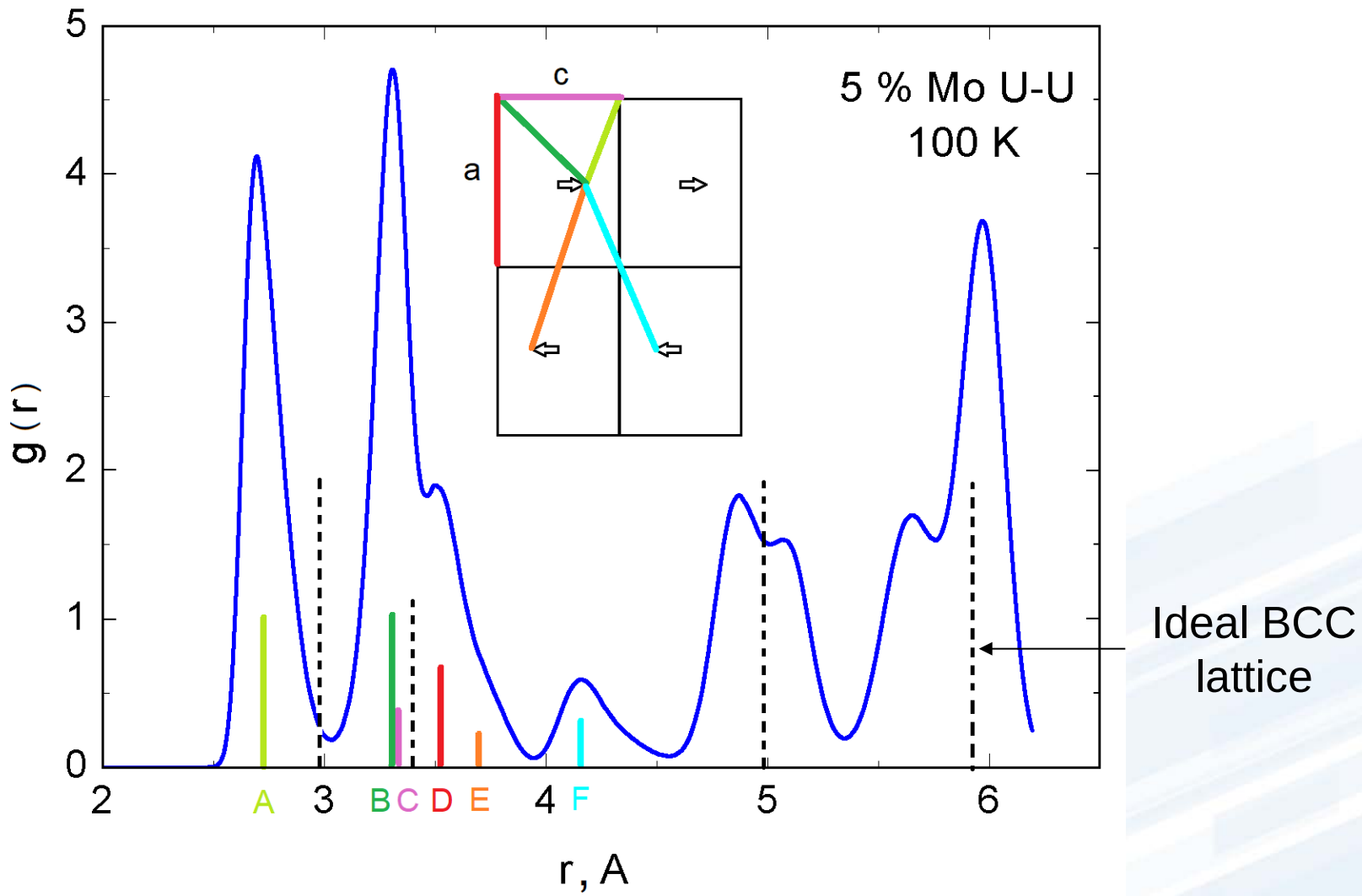
Radial distribution functions of γ^0 phase

— Ideal BCC lattice

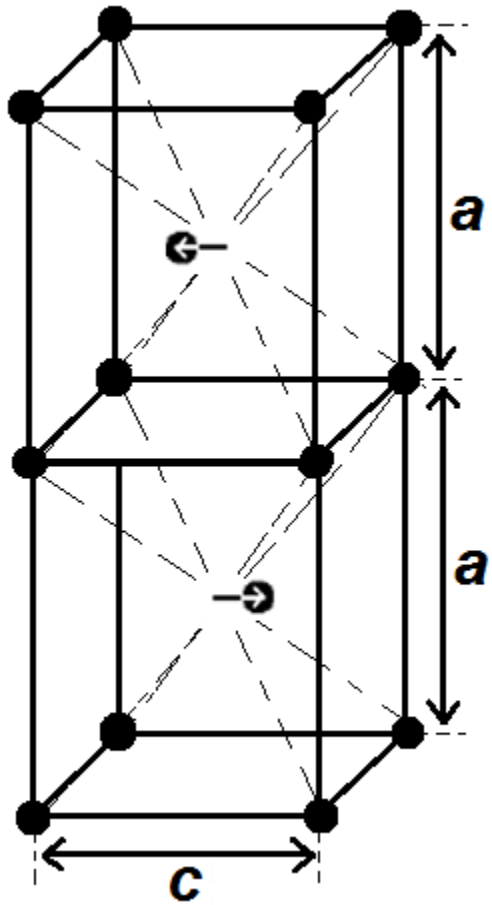




Radial distribution function of γ^0 phase

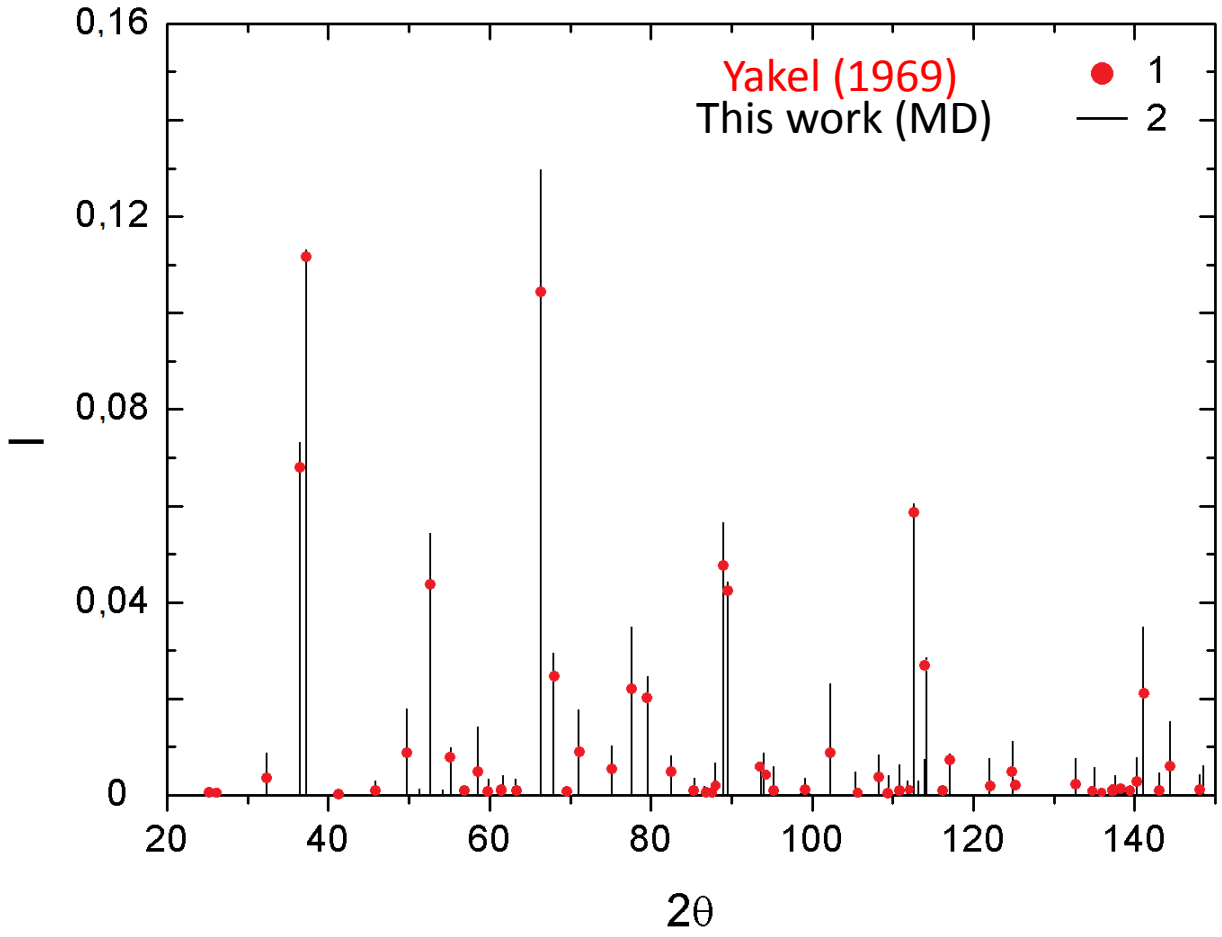


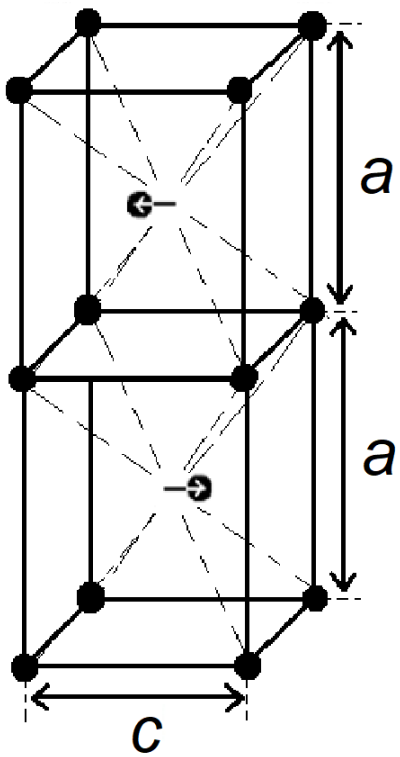
γ^0 structure



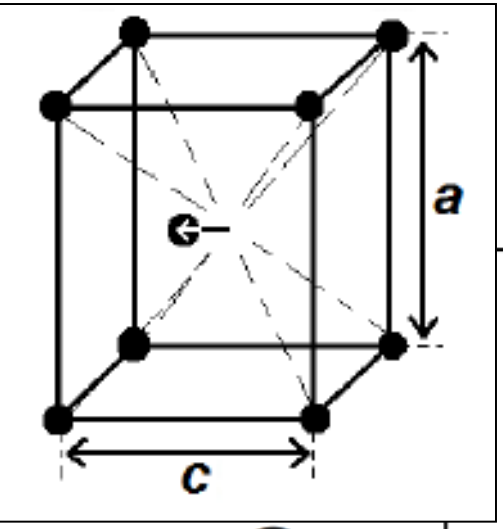
structure γ^0 phase is the same with Yakel's work (1969)

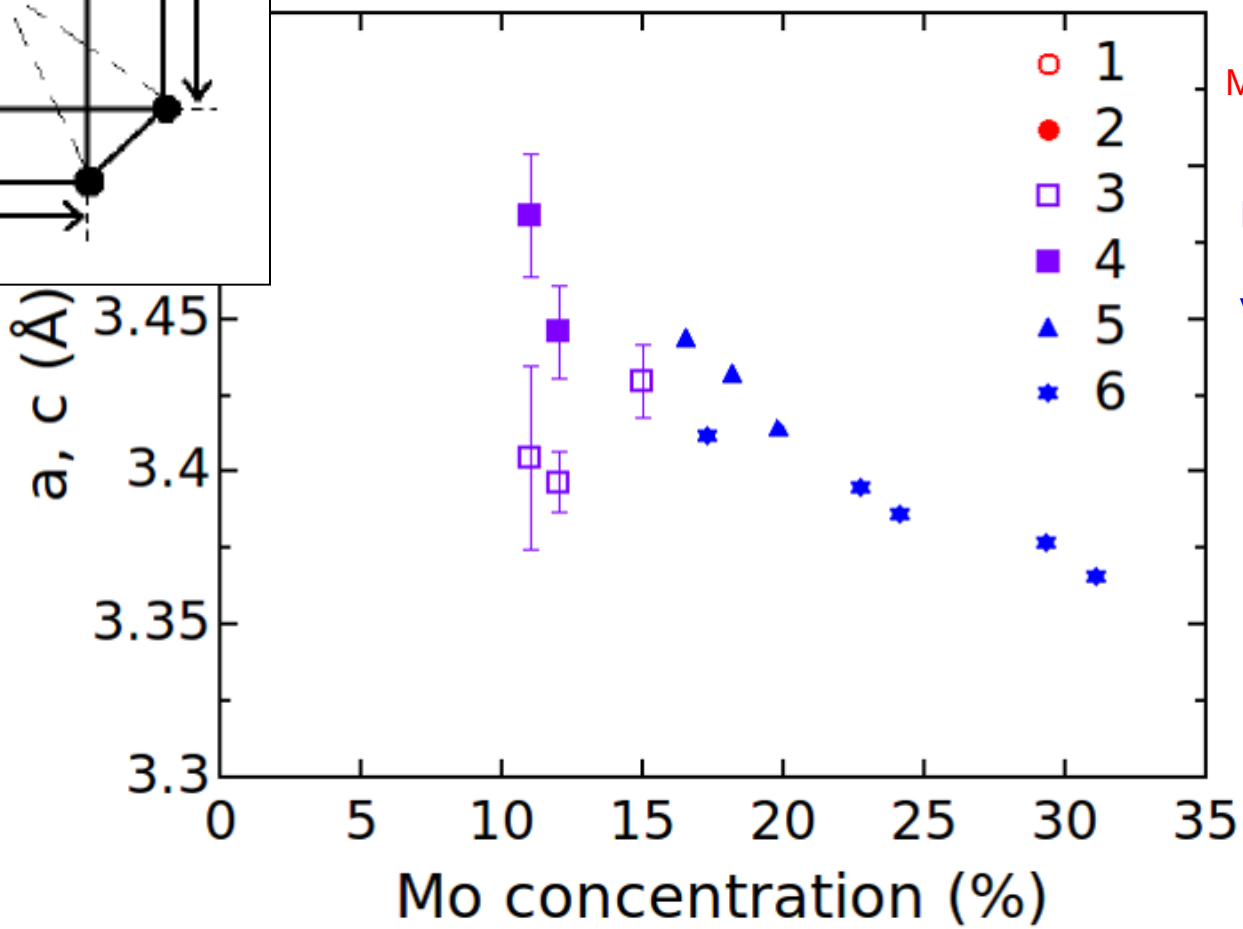
XRD of γ^0 phase





Anisotropy of γ^0





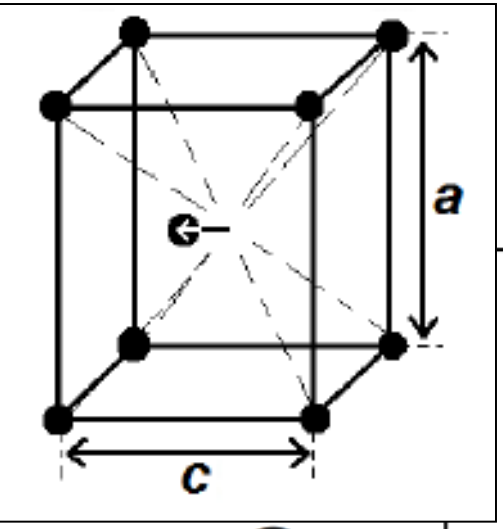
MD calculation, this work

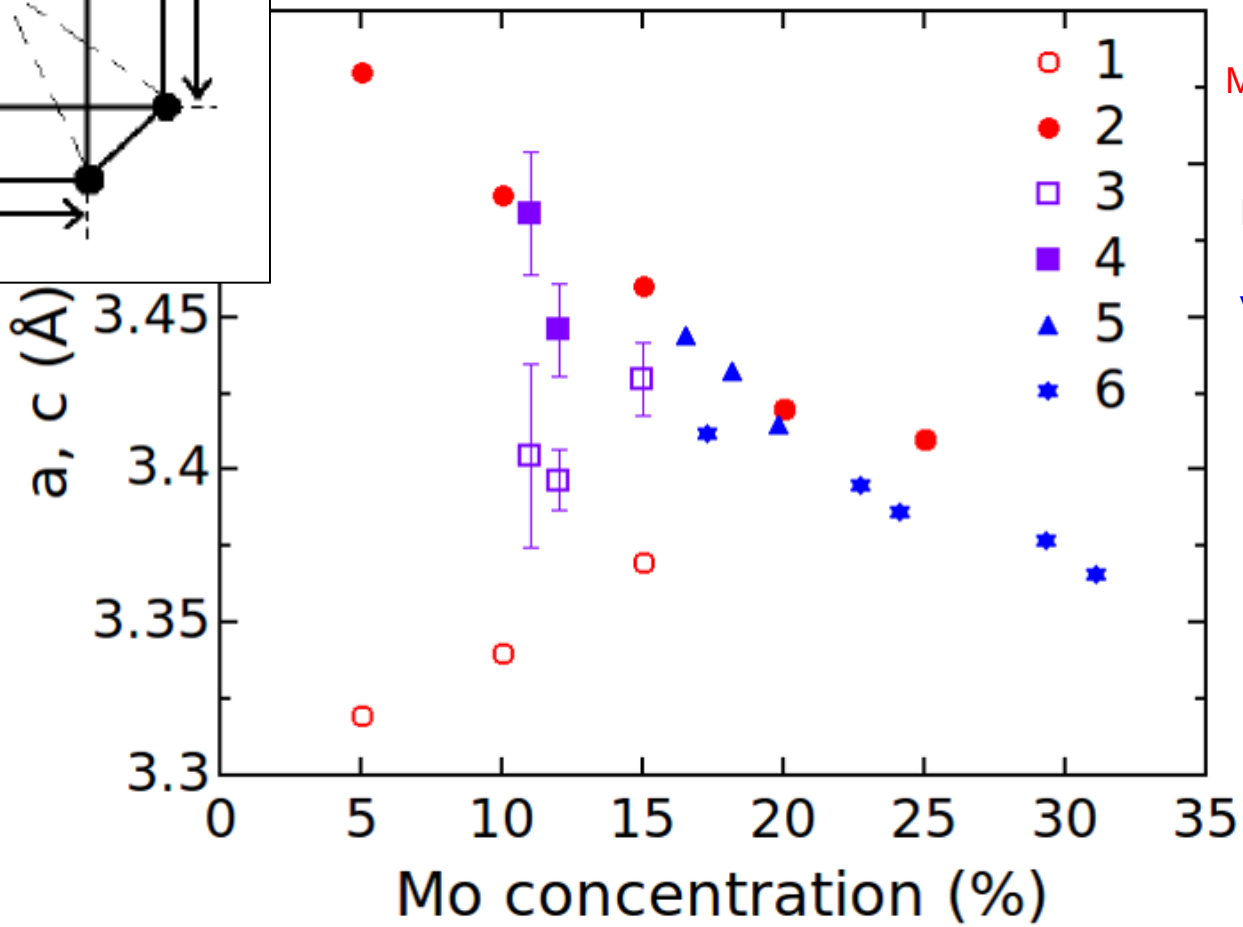
I. Tkach et al 2012

V.P. Sinha et al 2010

XRD database

Anisotropy of γ^0





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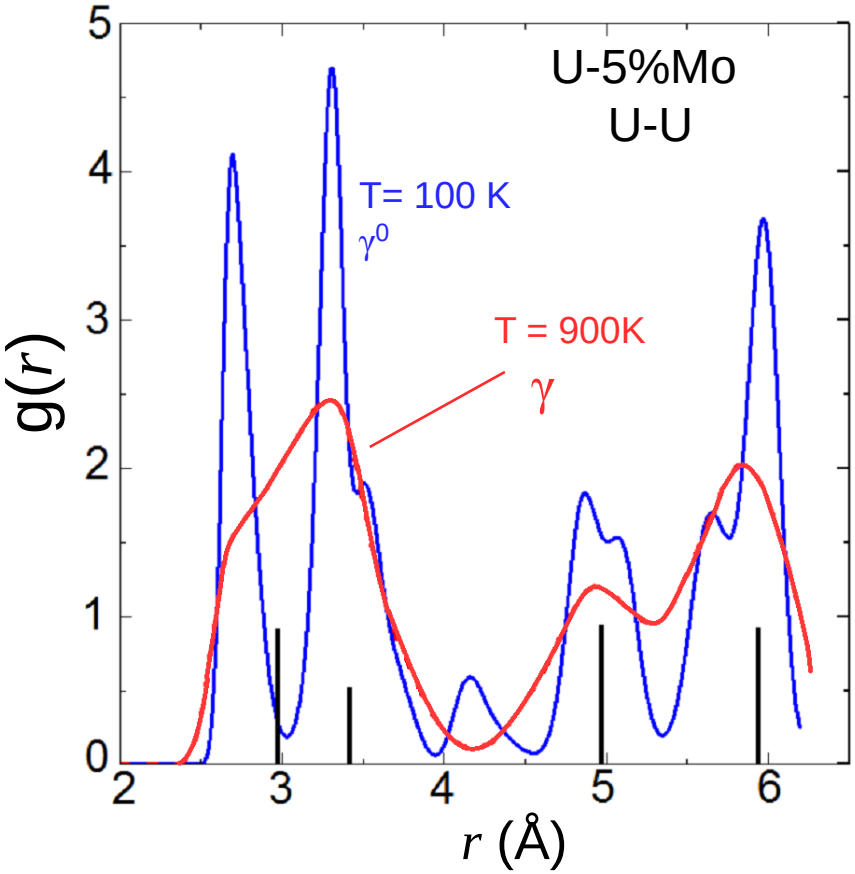
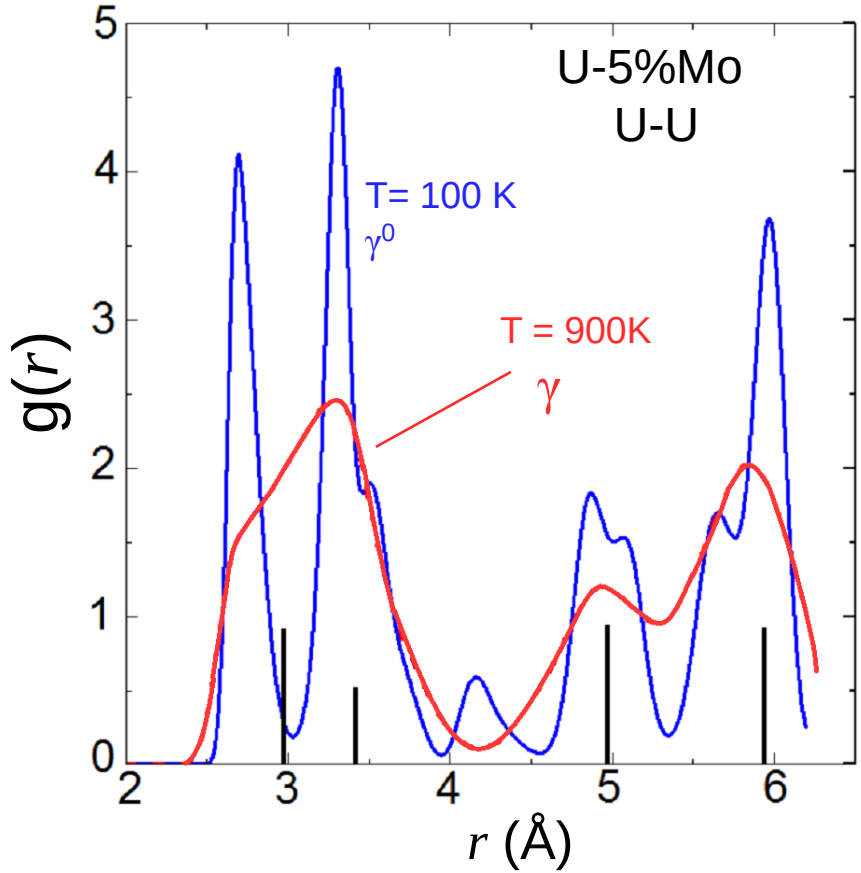
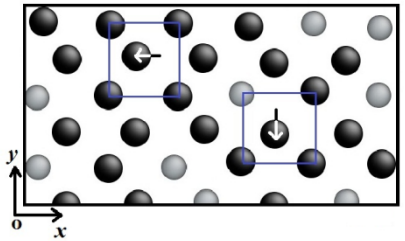


— Ideal BCC lattice

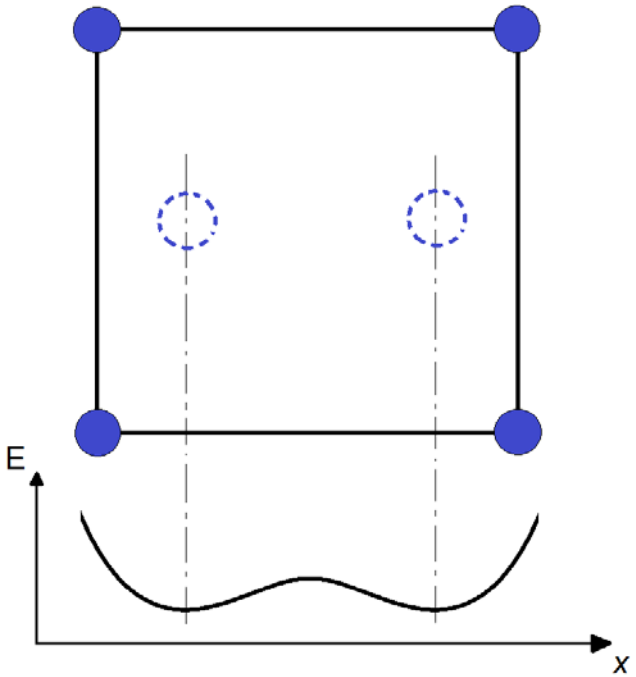
Radial distribution function of γ phase



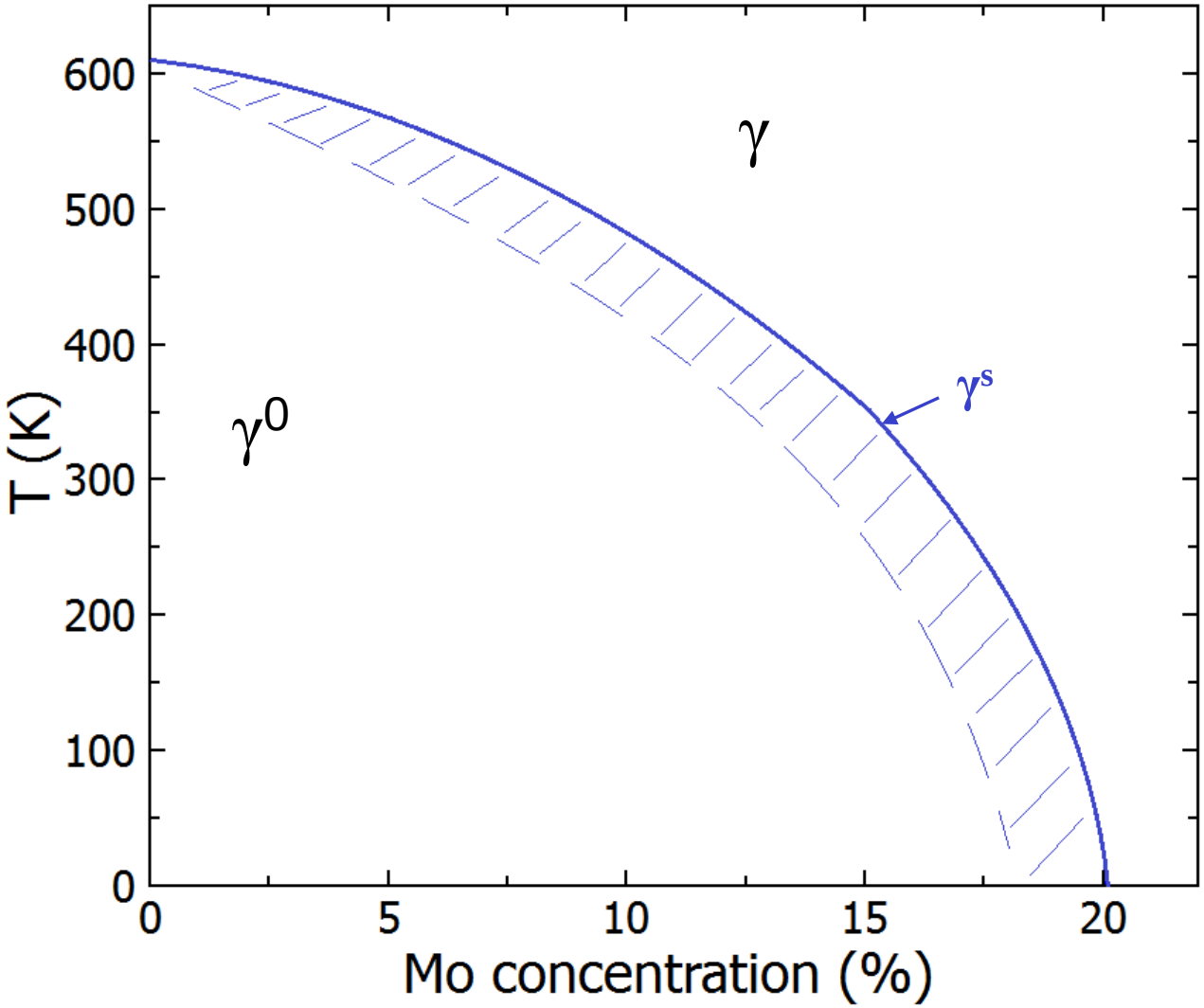
— Ideal BCC lattice



γ



Calculated areas of phase stability on T-x diagram for various structures



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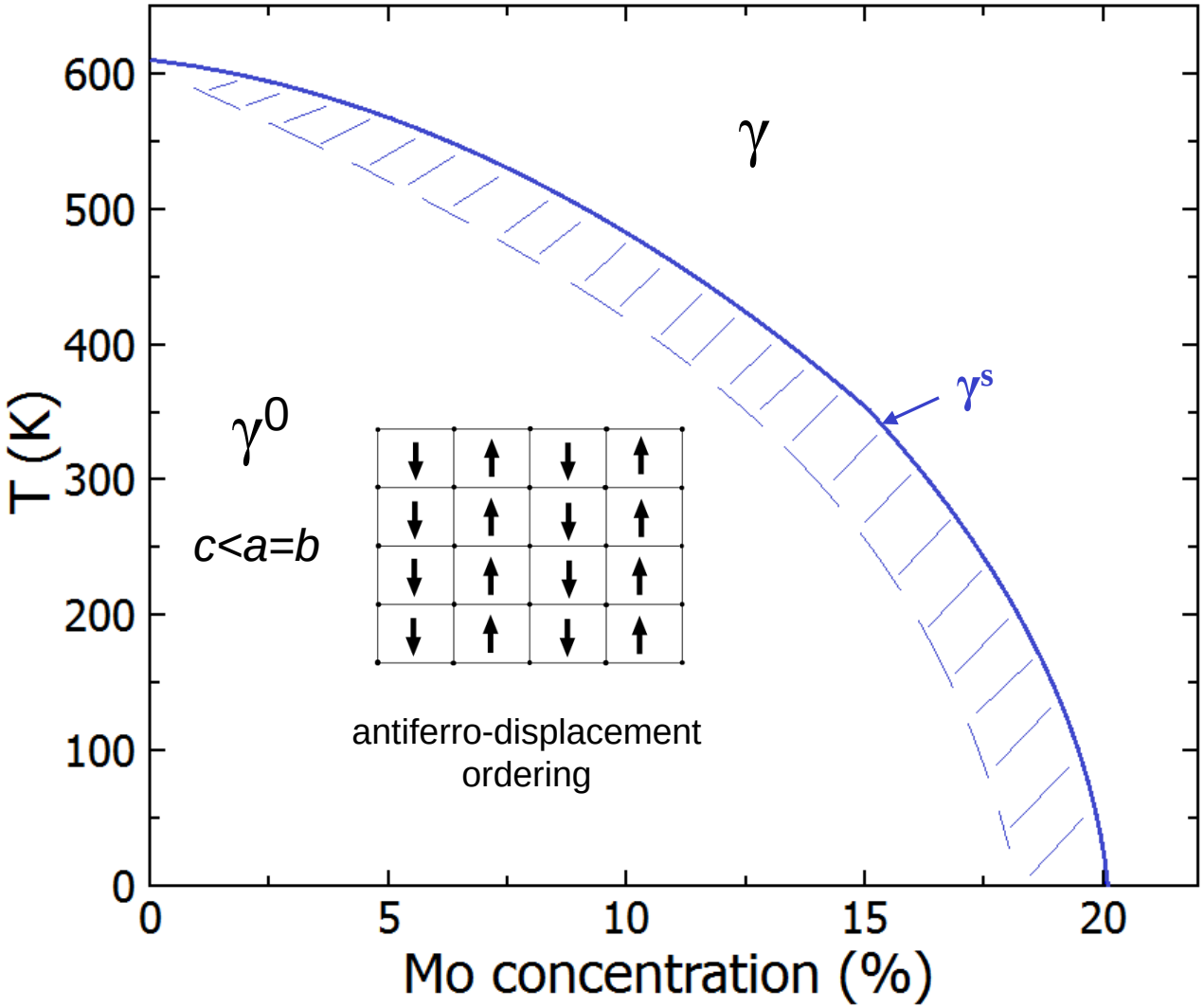

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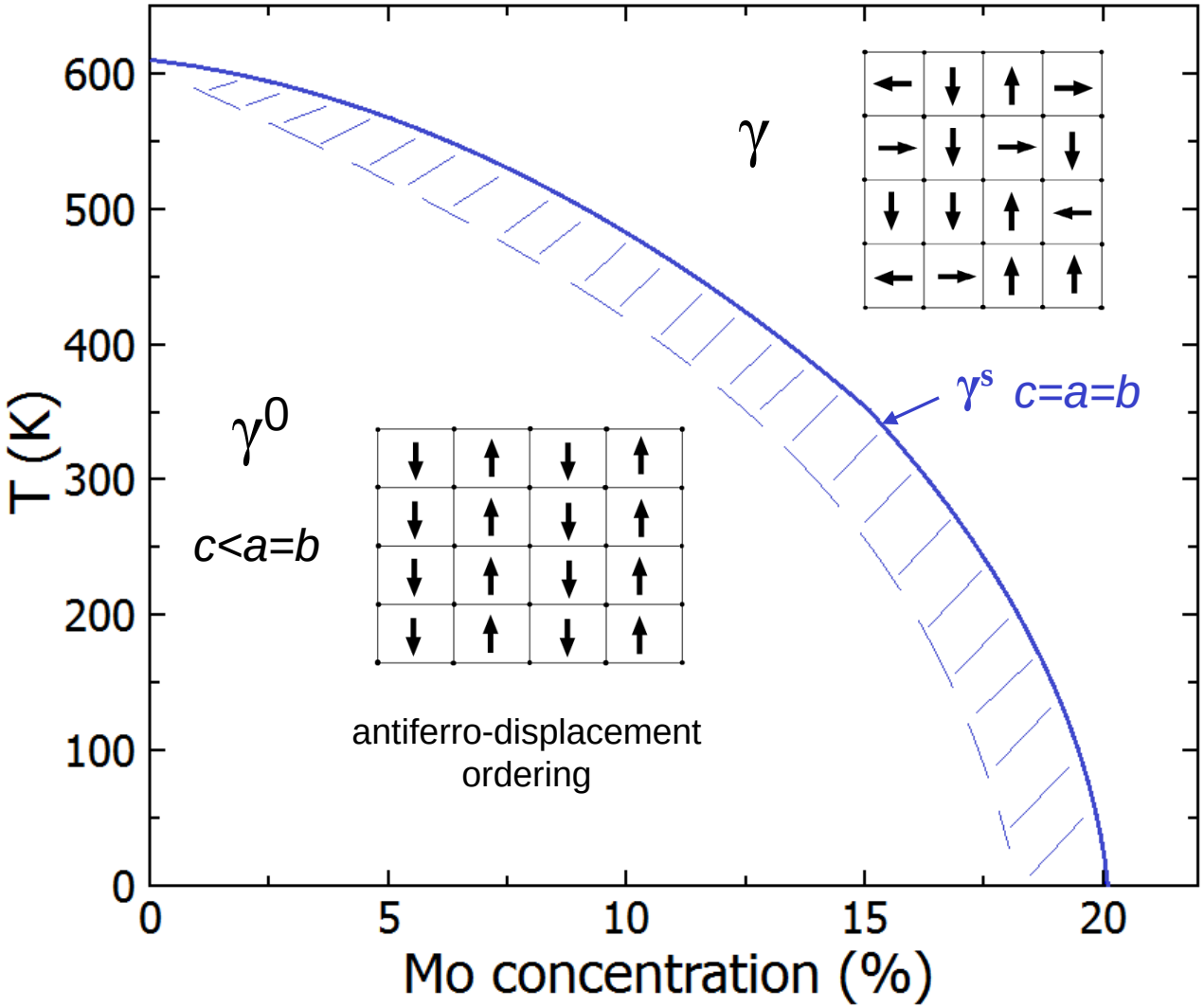

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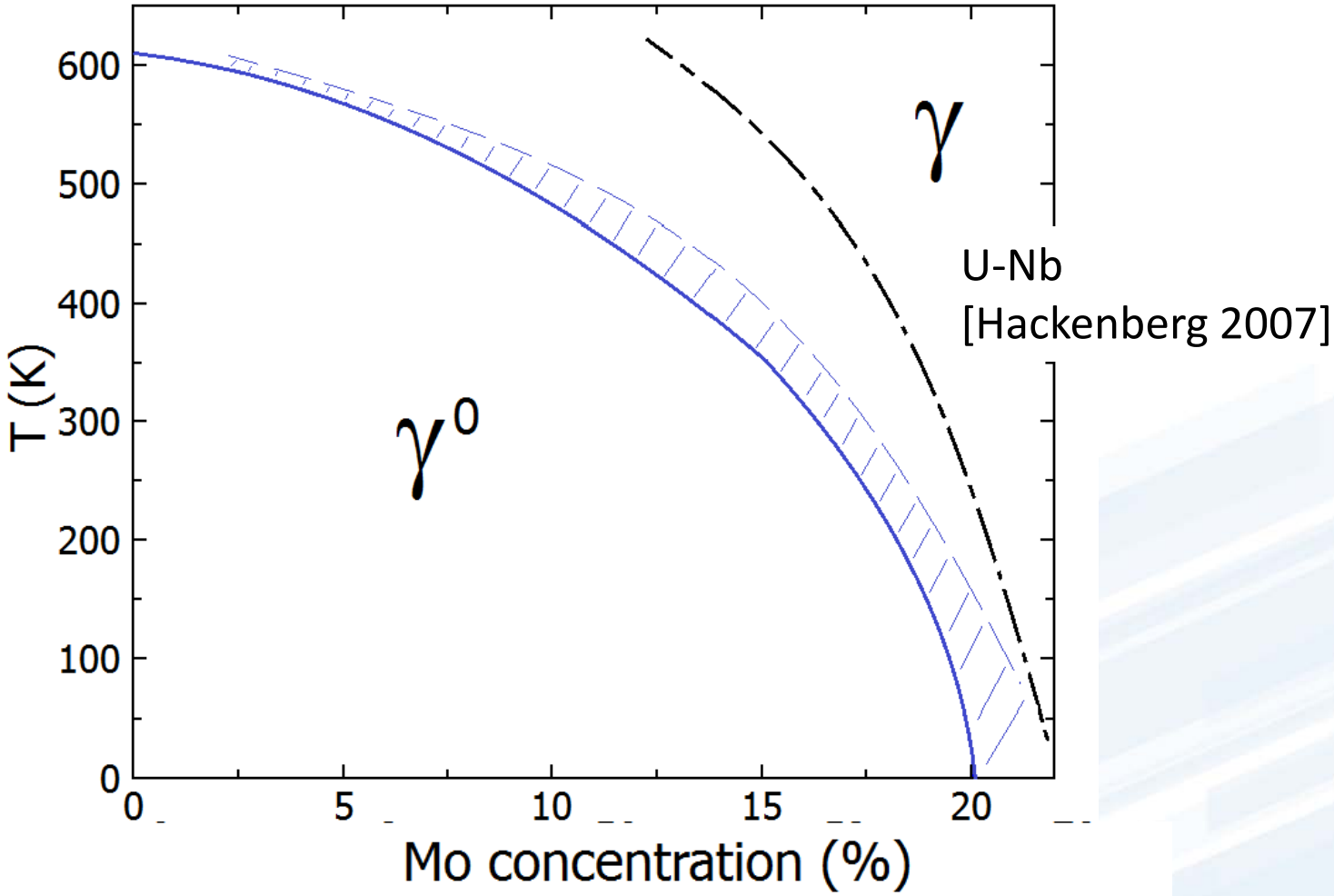
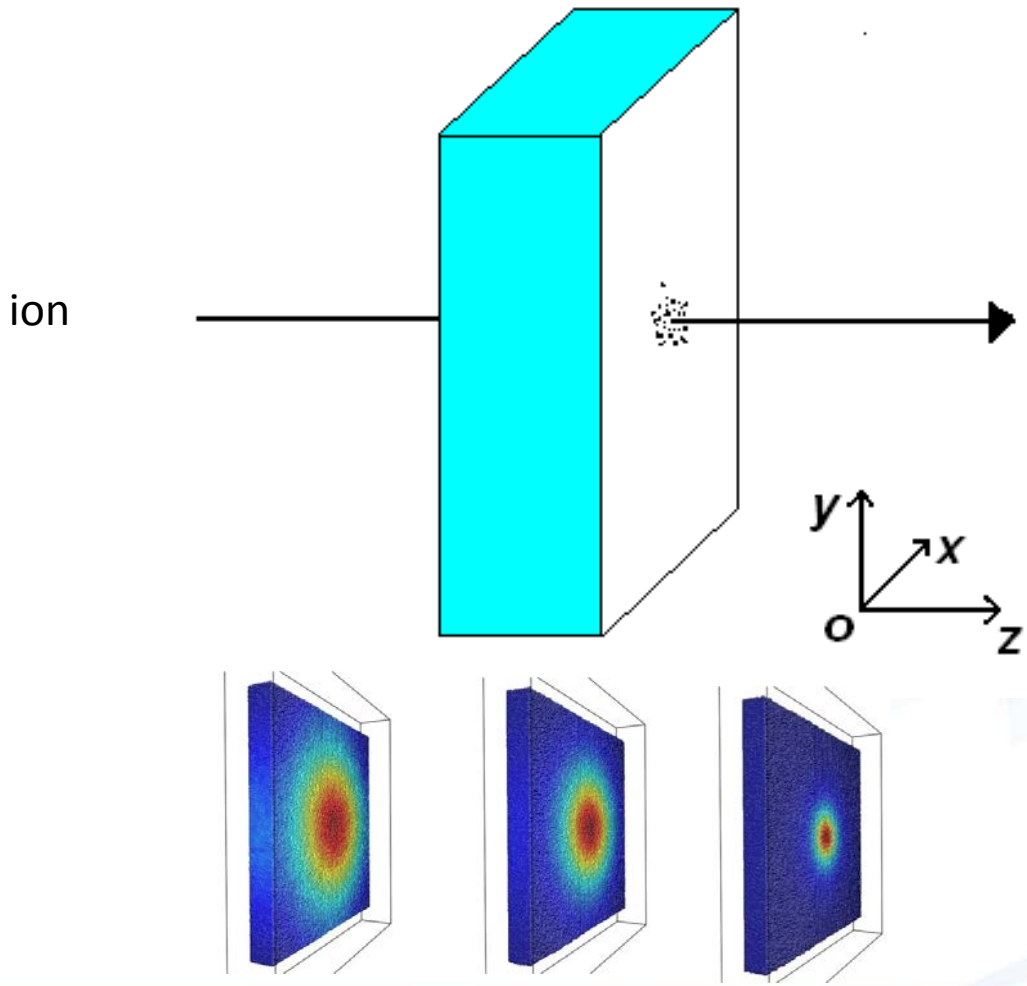


Structure transitions by irradiation effects

Moving of high energy ion through matter

E > 100 keV/n



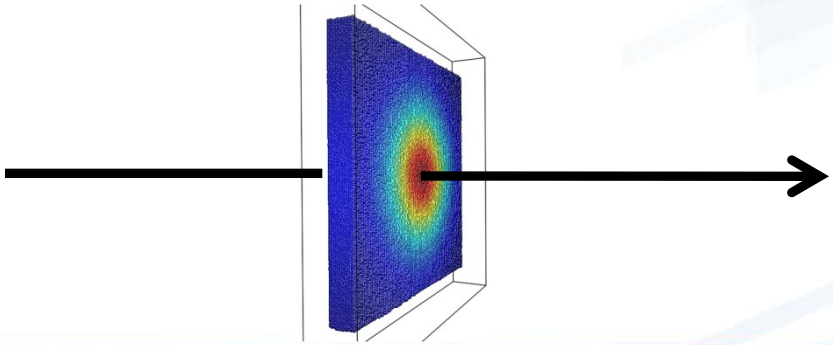
Thermal conductivity equation for ELECTRONS:

$$C_e \rho_e \frac{\partial T_e}{\partial t} = \nabla\left(\kappa_e \nabla T_e\right) - g_p\left(T_e - T_i\right)$$

Electron heat capacity

Electron thermal conductivity

Electron-ion relaxation



MD simulation for IONS :

$$\vec{F}_i = -\partial U / \partial \vec{r}_i + \vec{F}_{lang} - \nabla P_e / n_i$$

Interionic potential

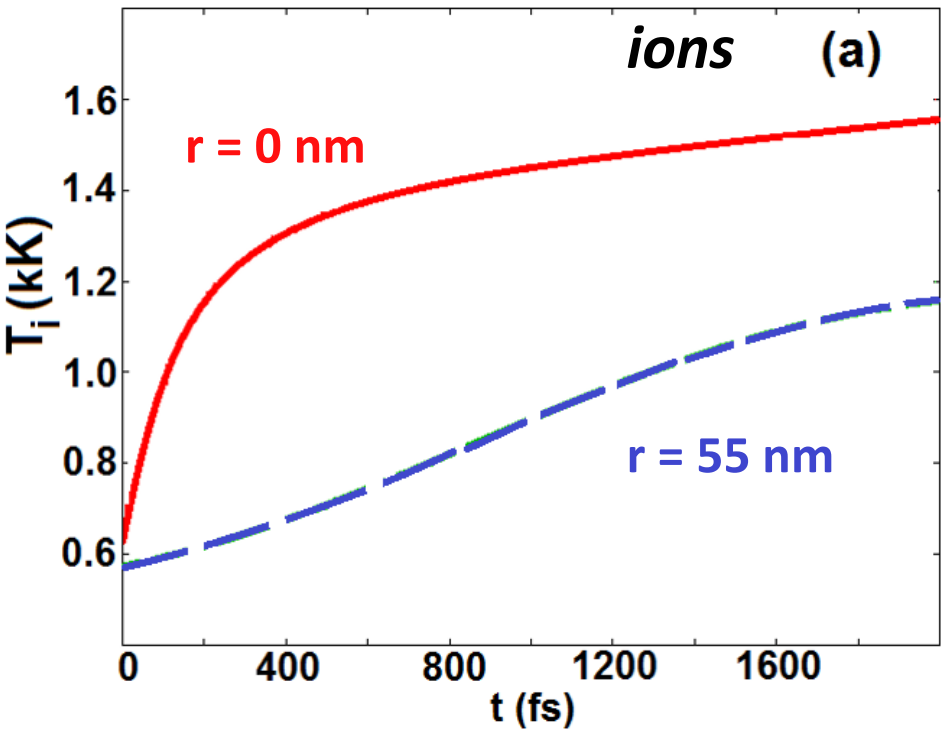
Electron-ion relaxation Langevin thermostat

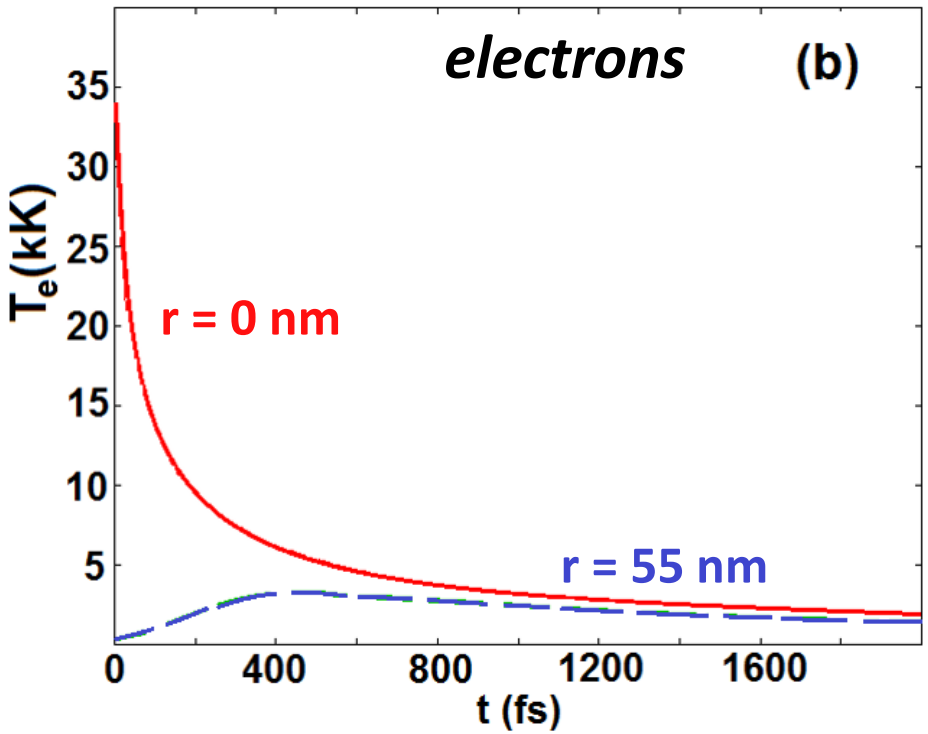
Electron pressure

Norman G E, Starikov S V, Stegailov V V et al. 2013 *Contrib. Plasma Phys.* **53** 129-139
Pisarev V V and Starikov S V 2014 *J. Phys.: Condens. Matter* **26** 475401

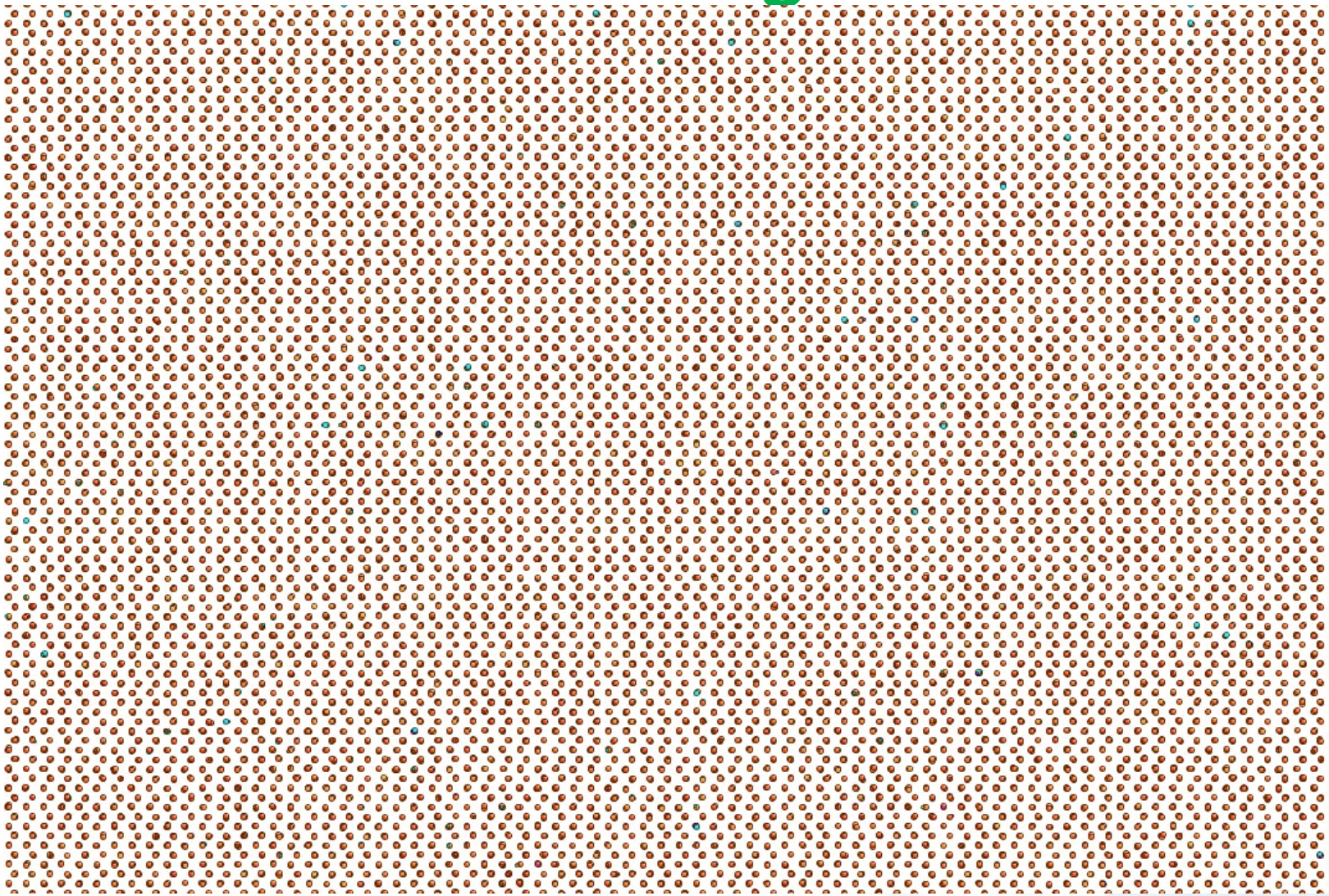
Thermal spike model

dE/dz = 27 keV/nm

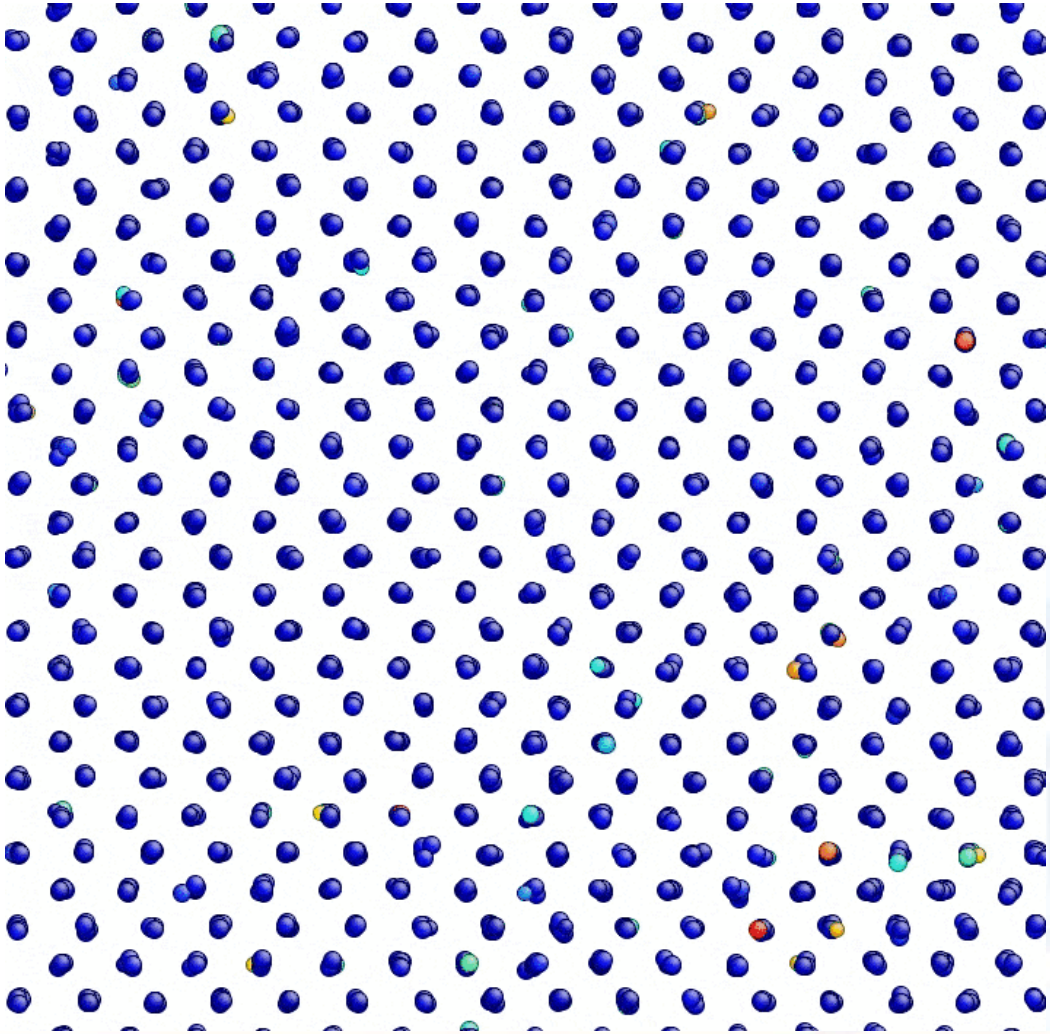




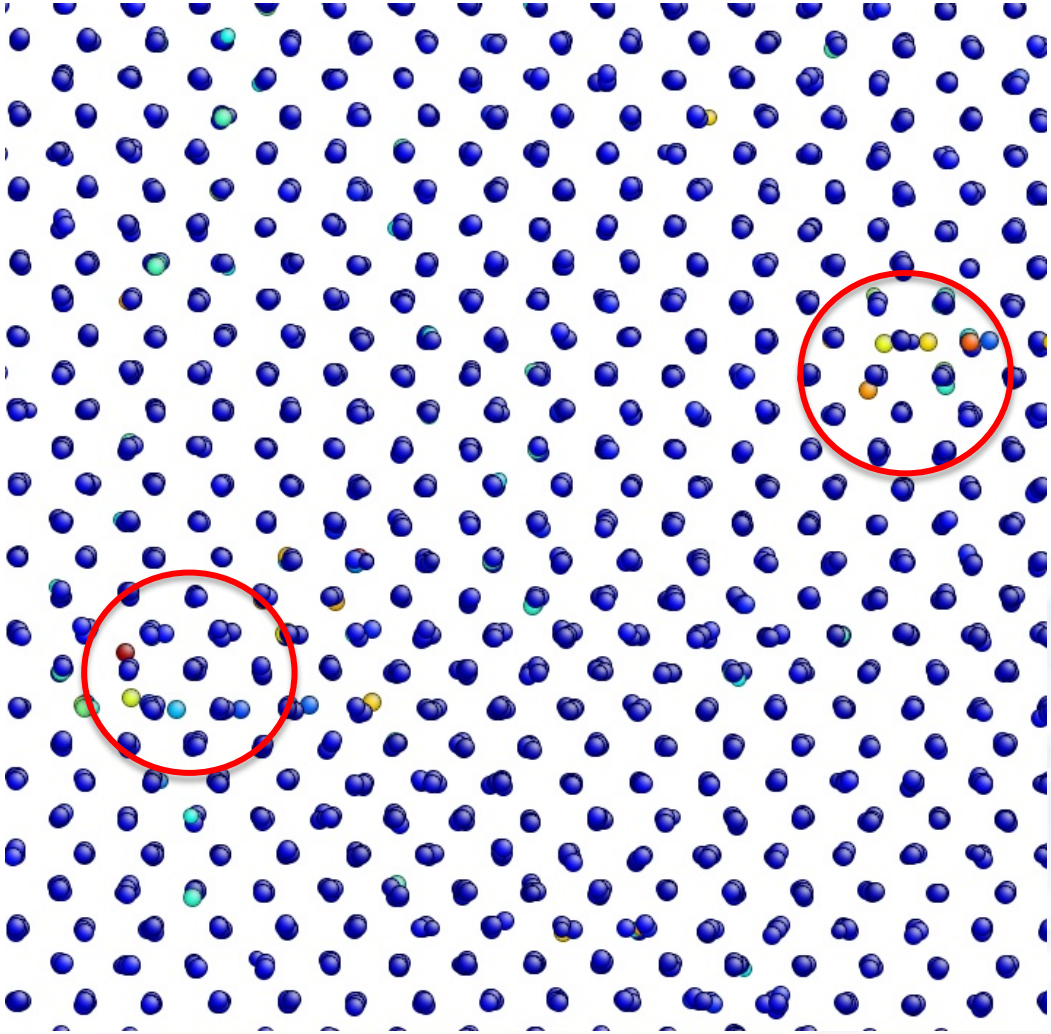
Radiation defects formation after melting



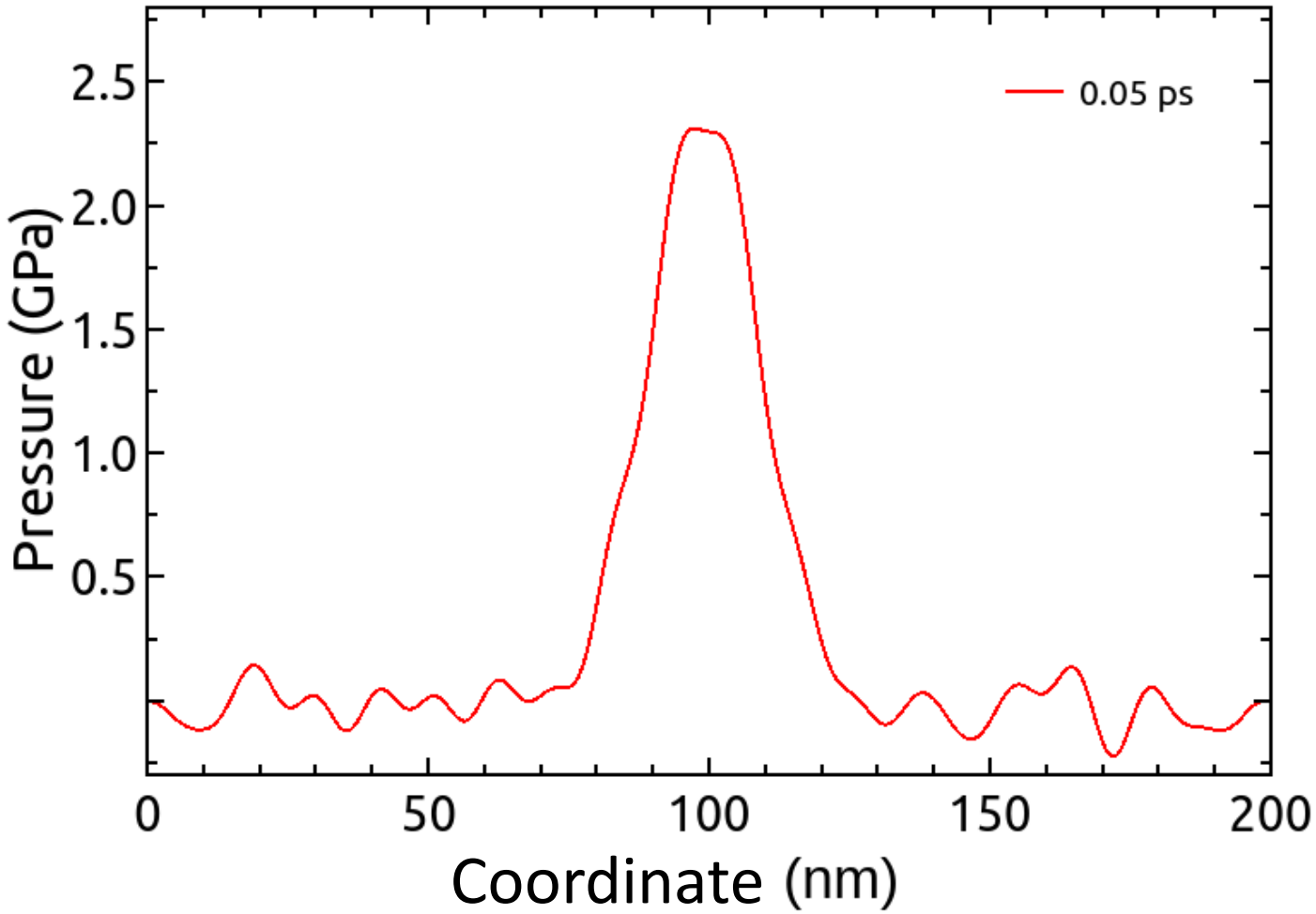
Radiation defects formation without melting



Radiation defects formation without melting



Pressure profiles along the radial direction to the axis of the track



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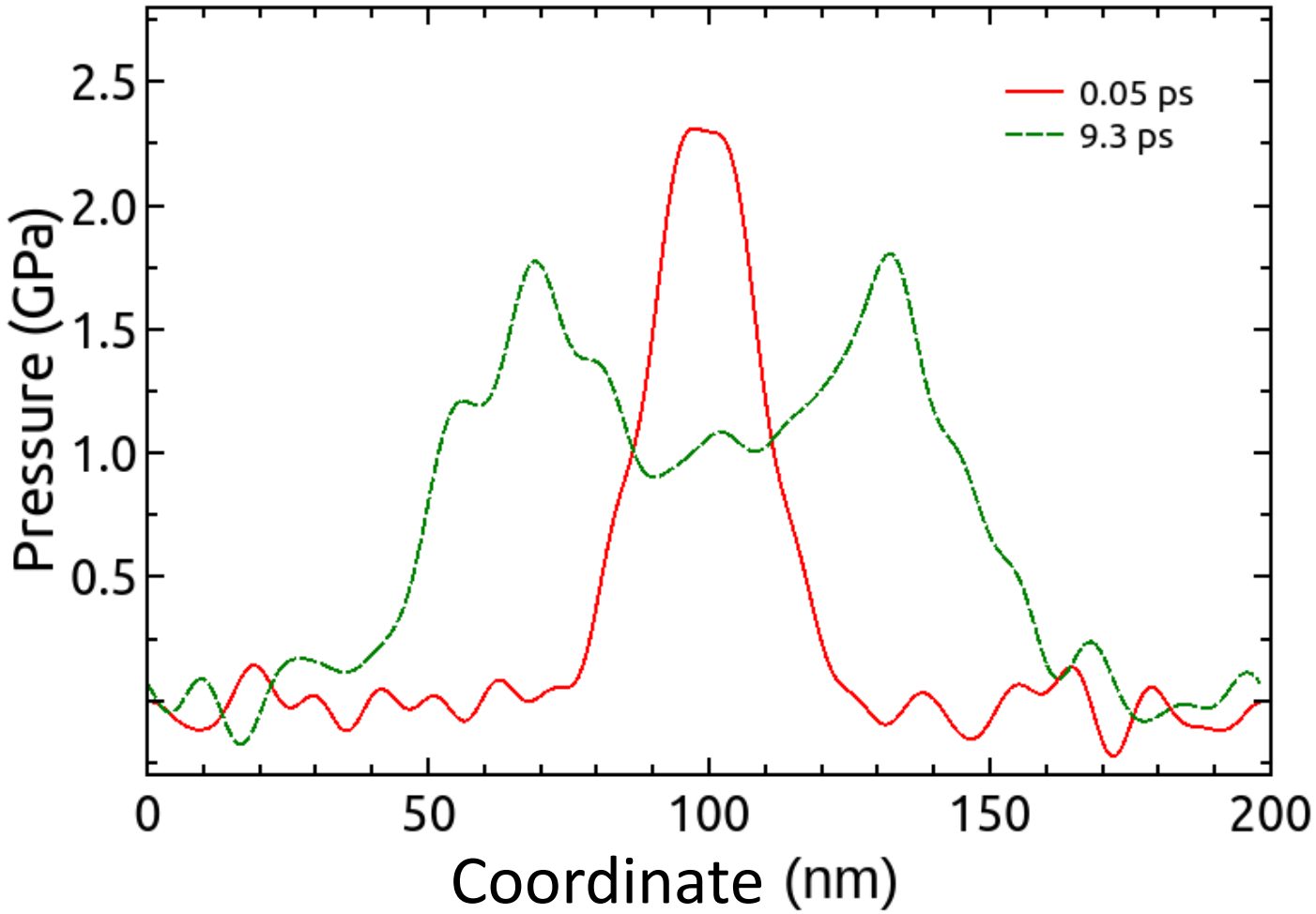

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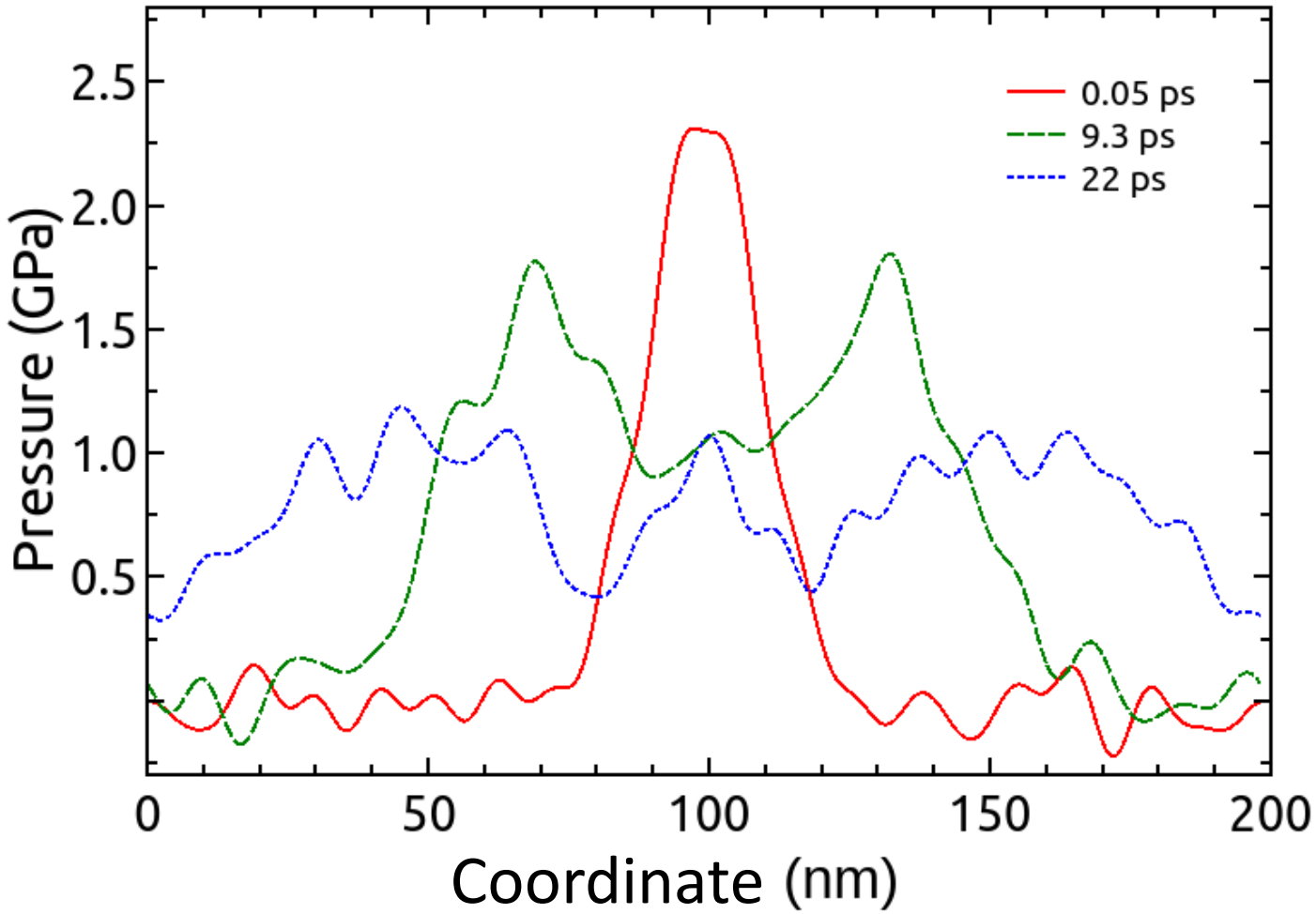
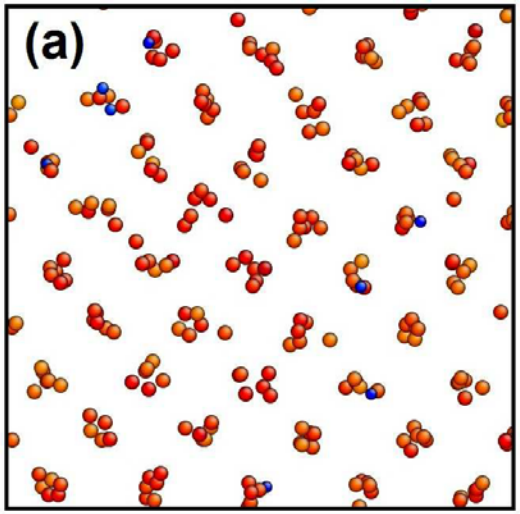


Formation of radiation defects in the atomistic simulation



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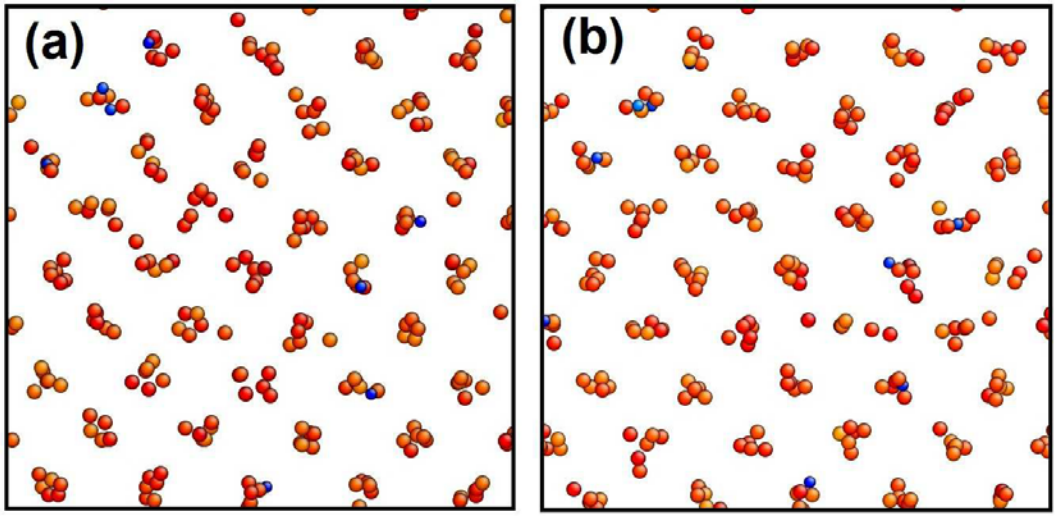

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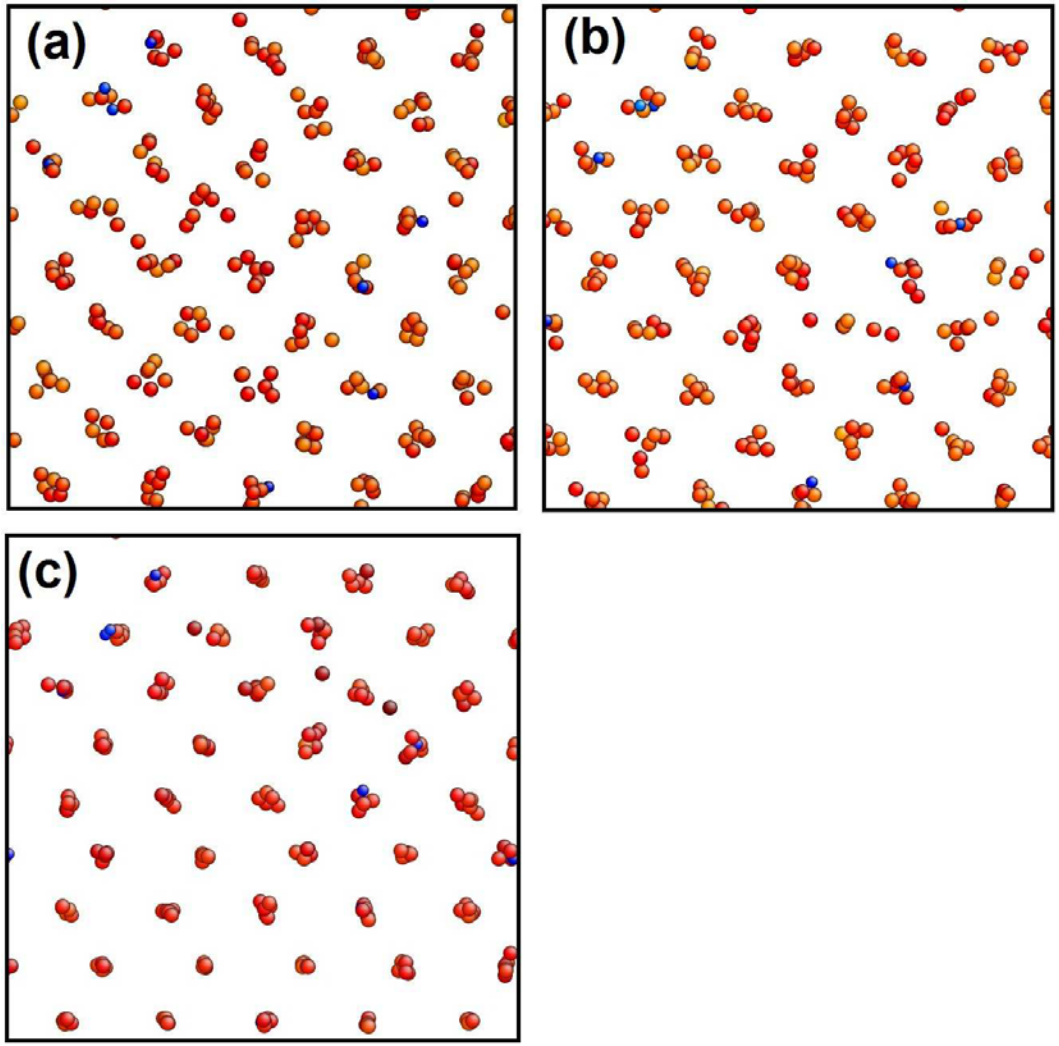
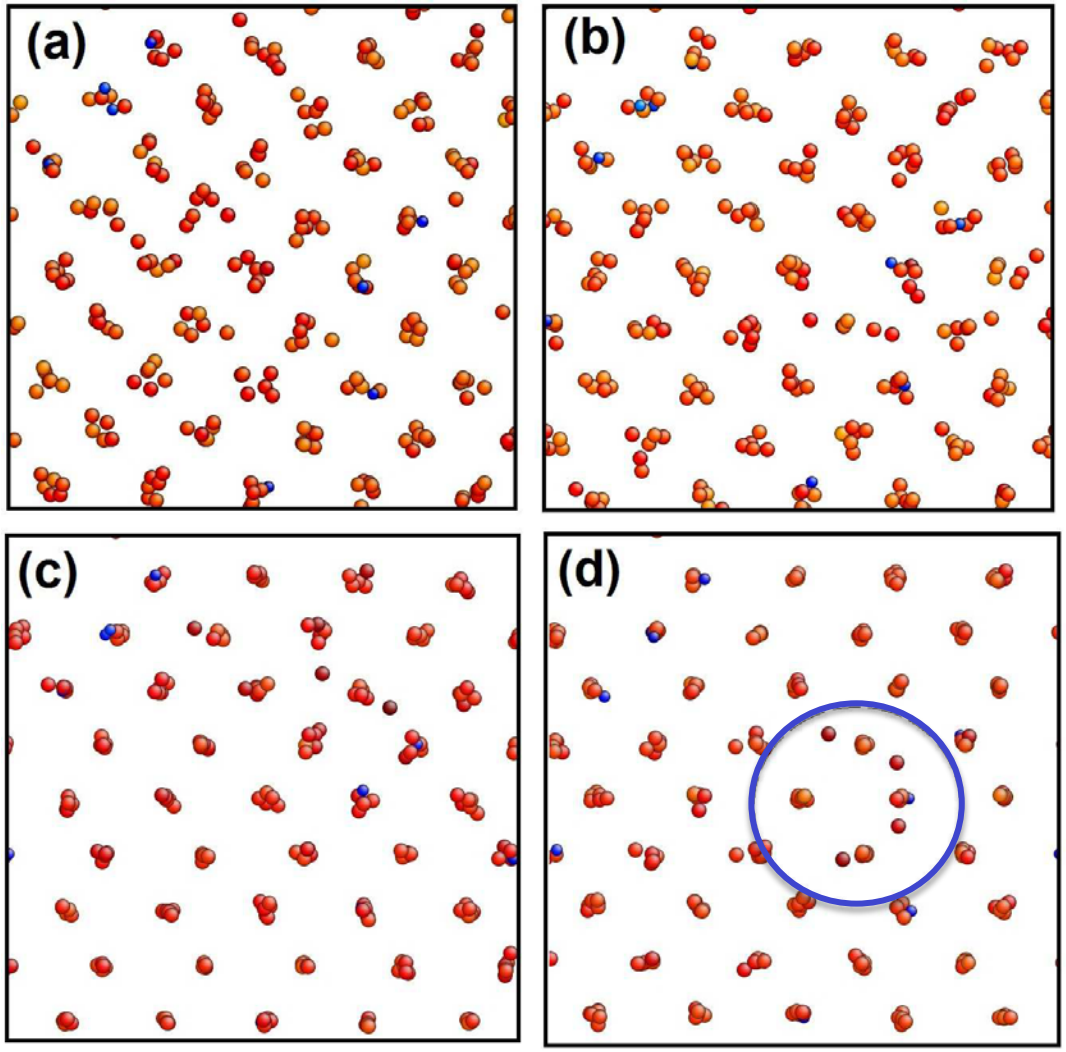
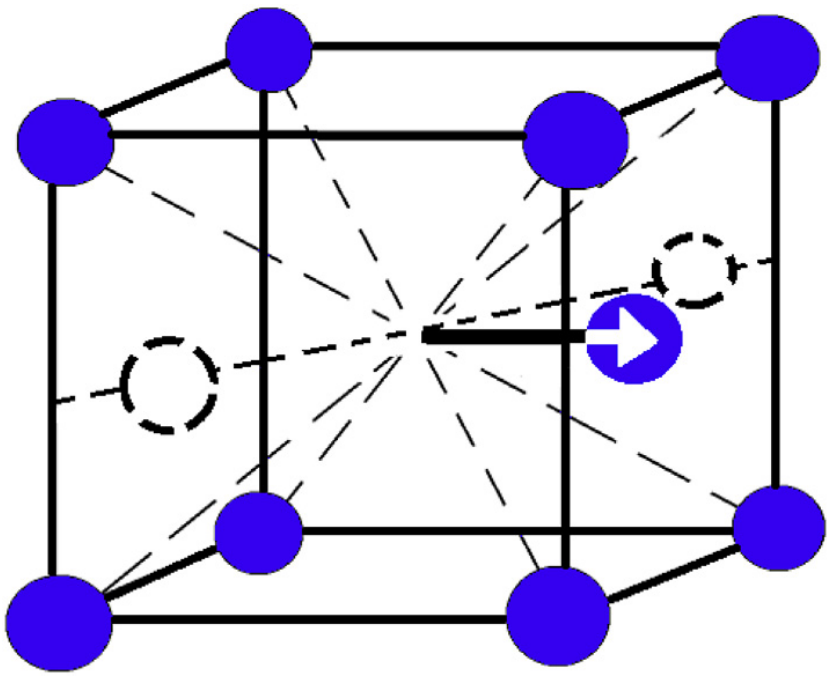


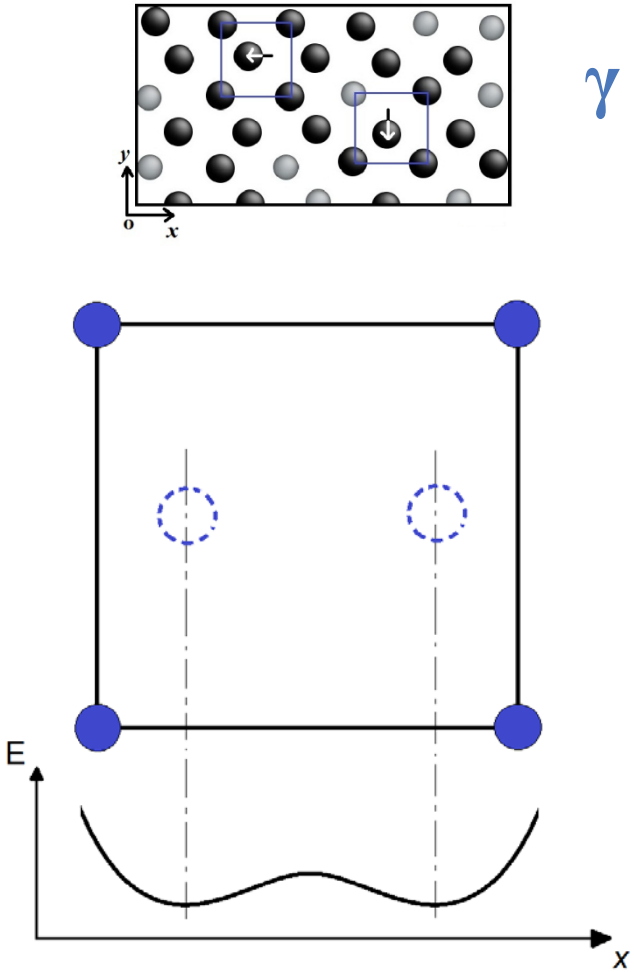
Formation of radiation defects in the atomistic simulation



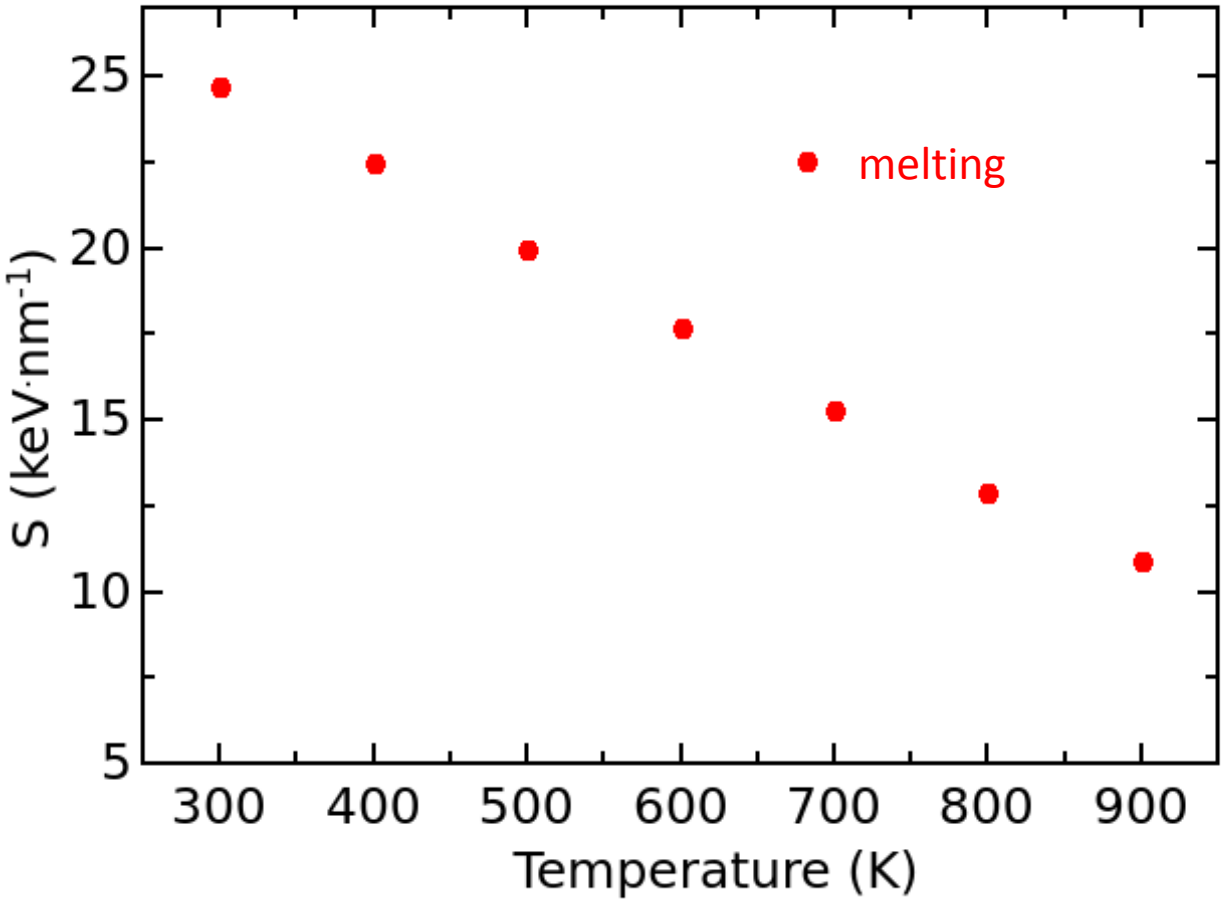
High temperature U-Mo alloy γ structure



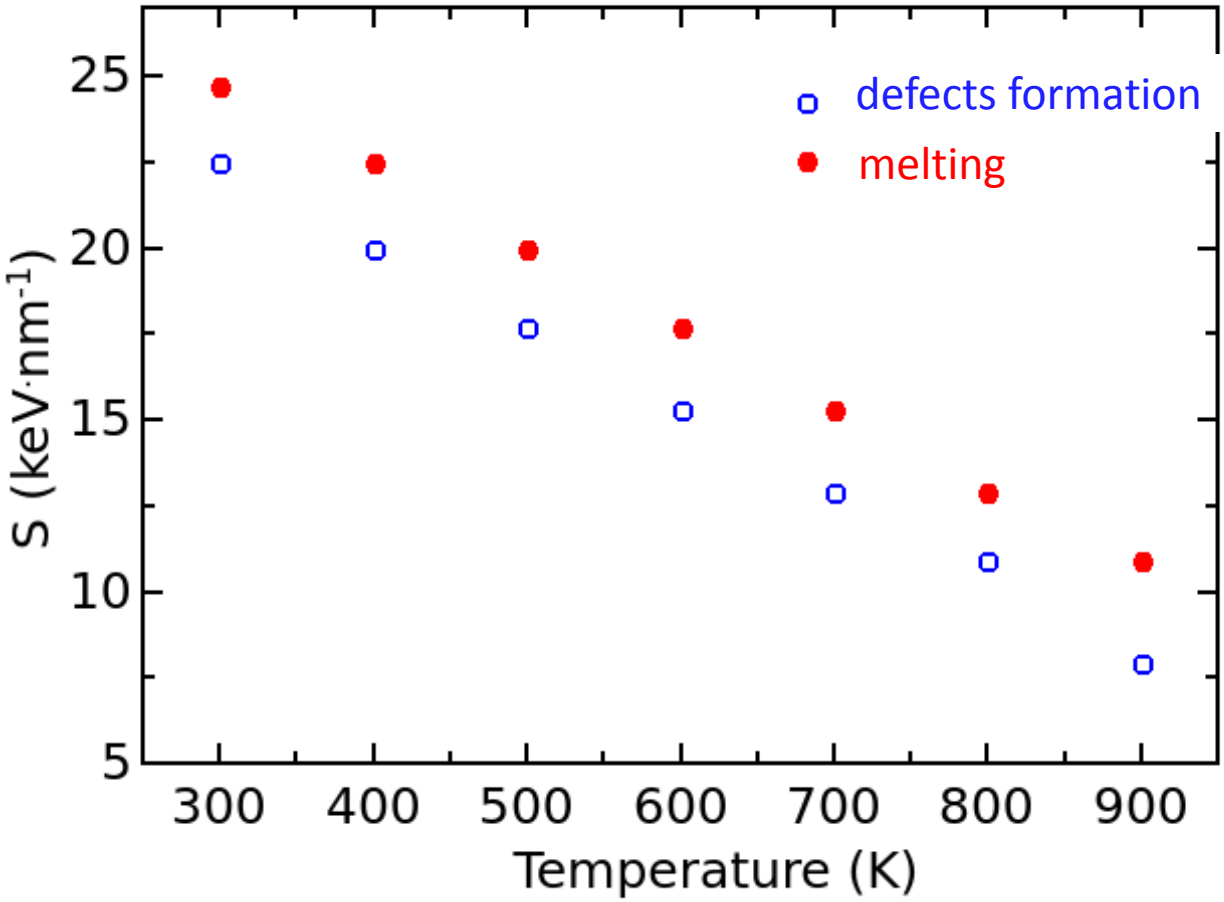
Empty atoms show position of SIA in quasi-bcc lattice of γ-phase (basic cell is shown by blue solid atoms).



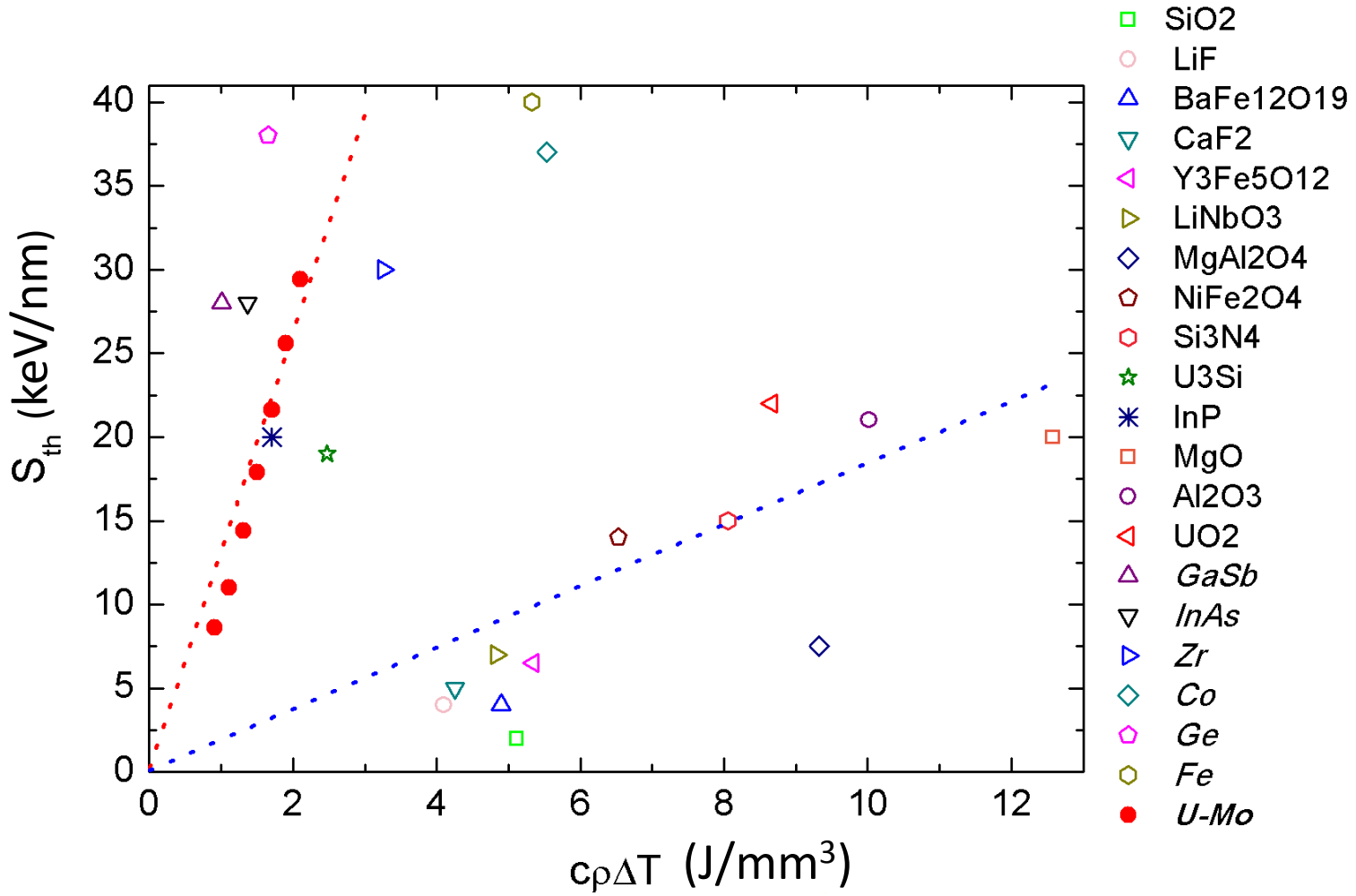
The threshold stopping power of swift ions dependence on temperature



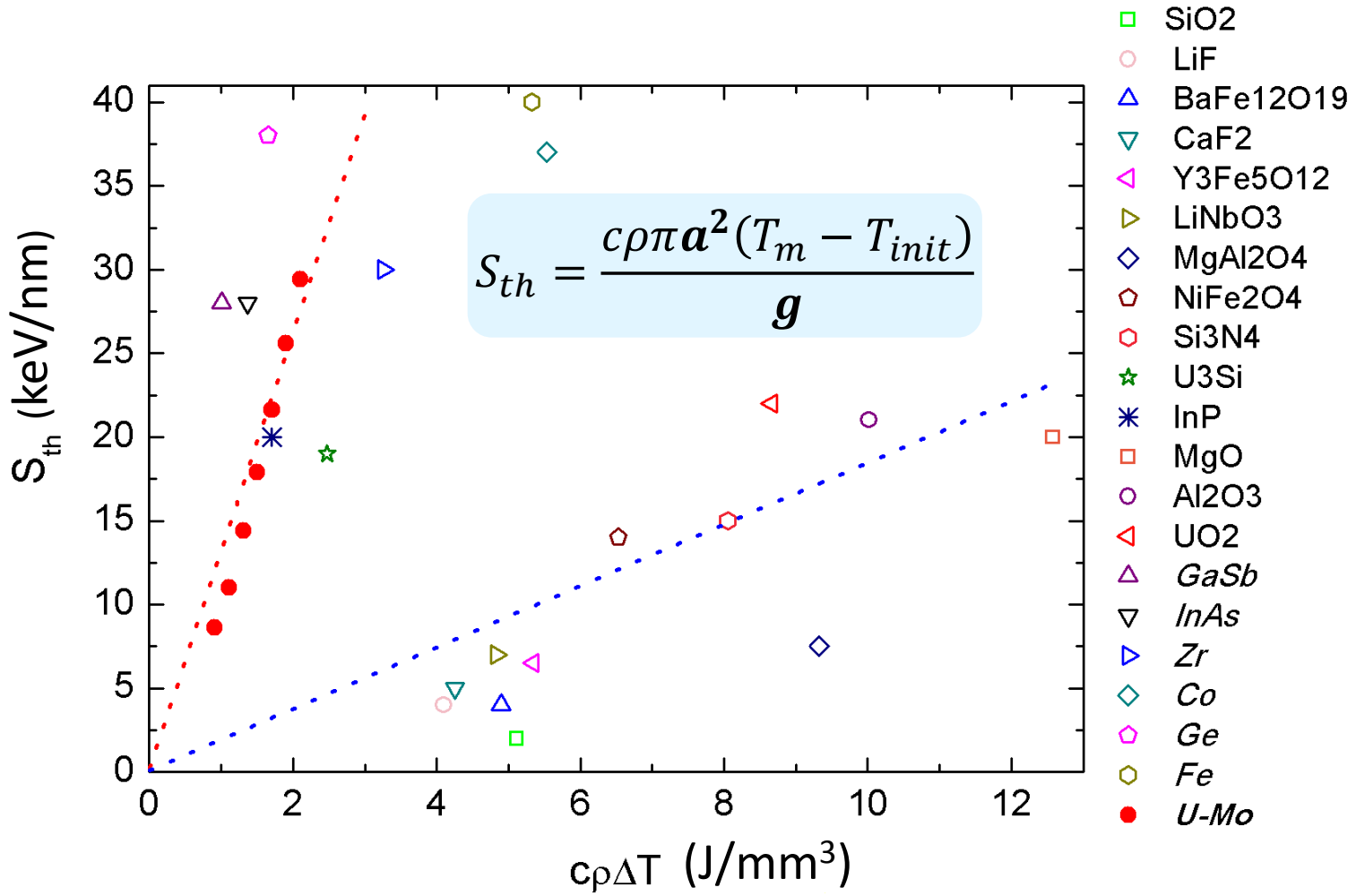
The threshold stopping power of swift ions dependence on temperature



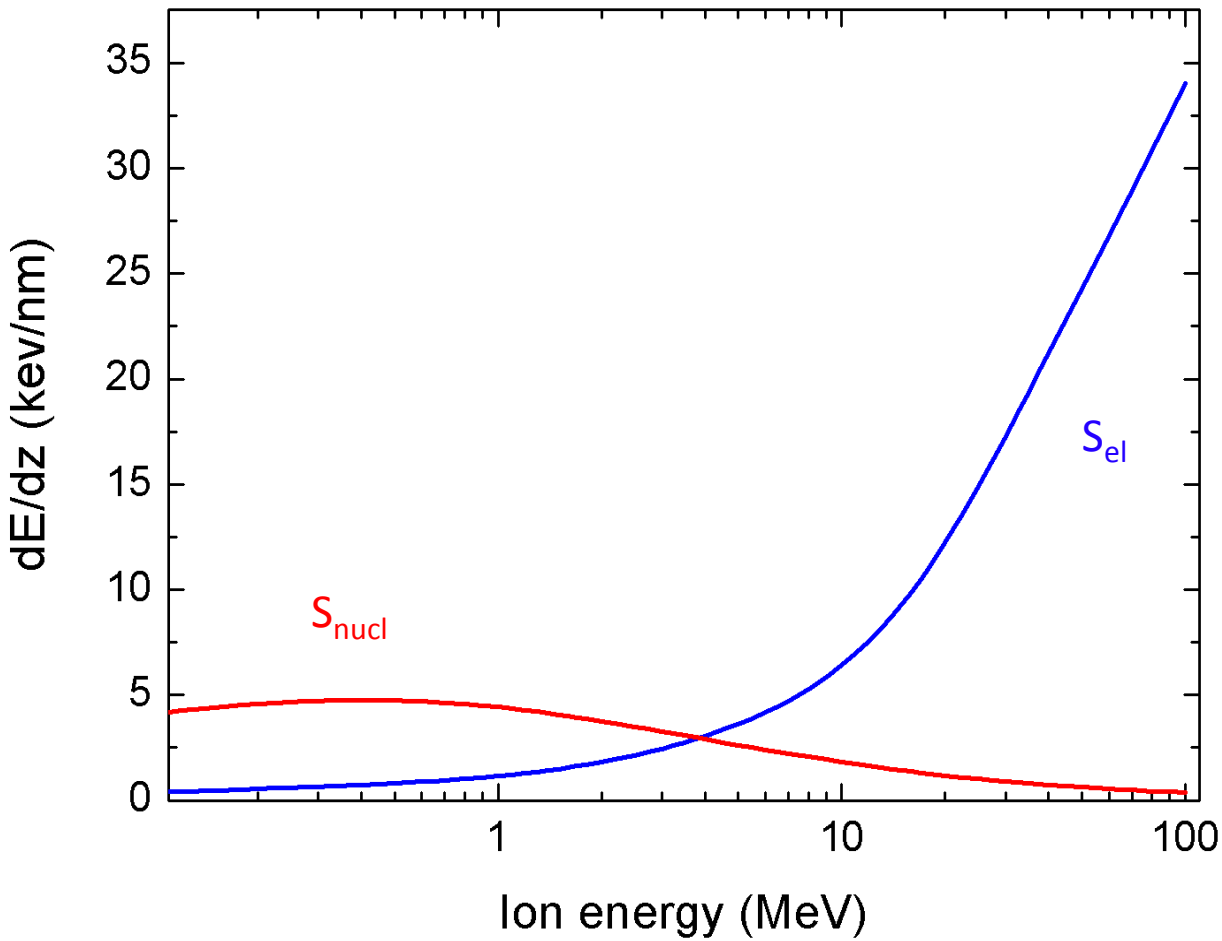
The threshold stopping power of swift ions dependence on temperature



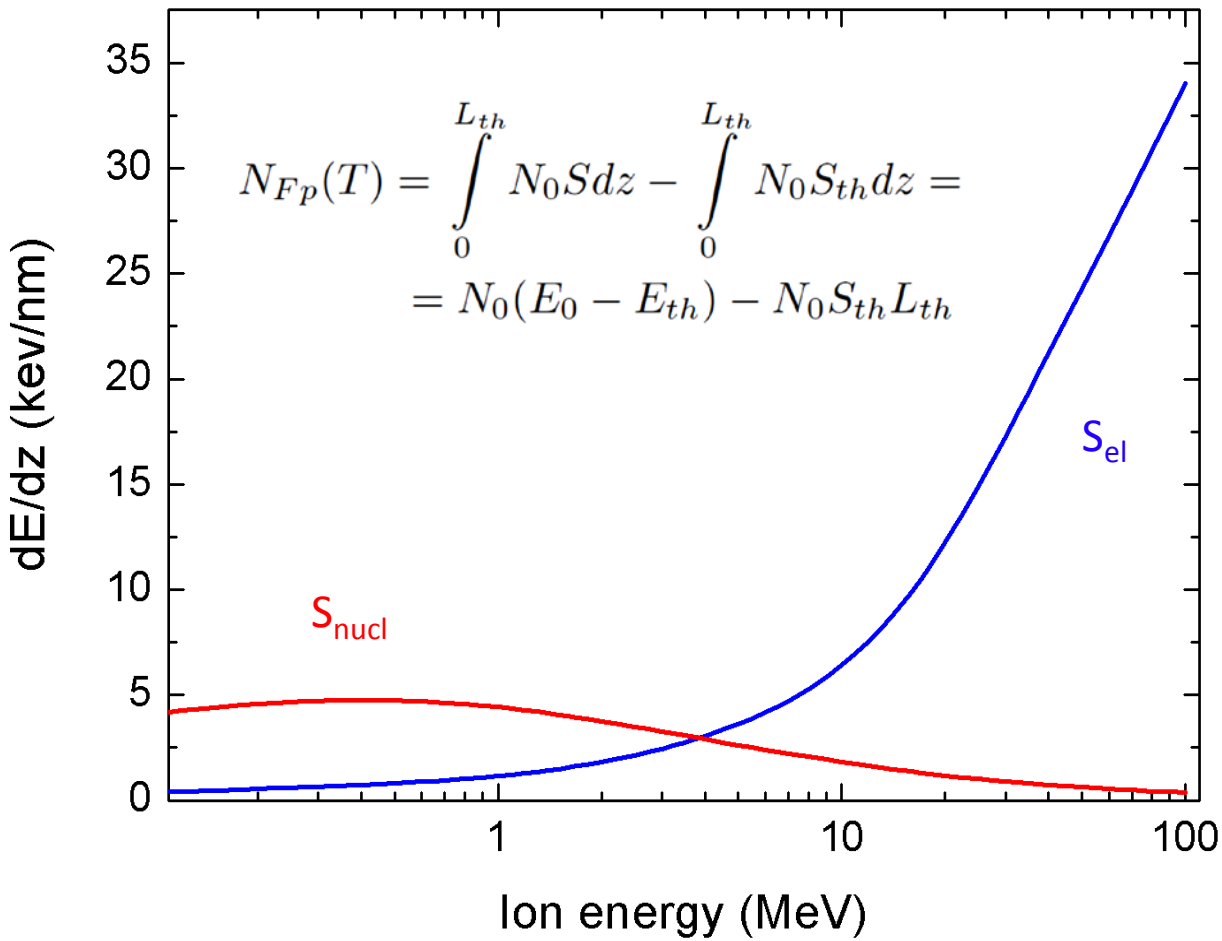
The threshold stopping power of swift ions dependence on temperature



The number of Frenkel pairs dependence on temperature



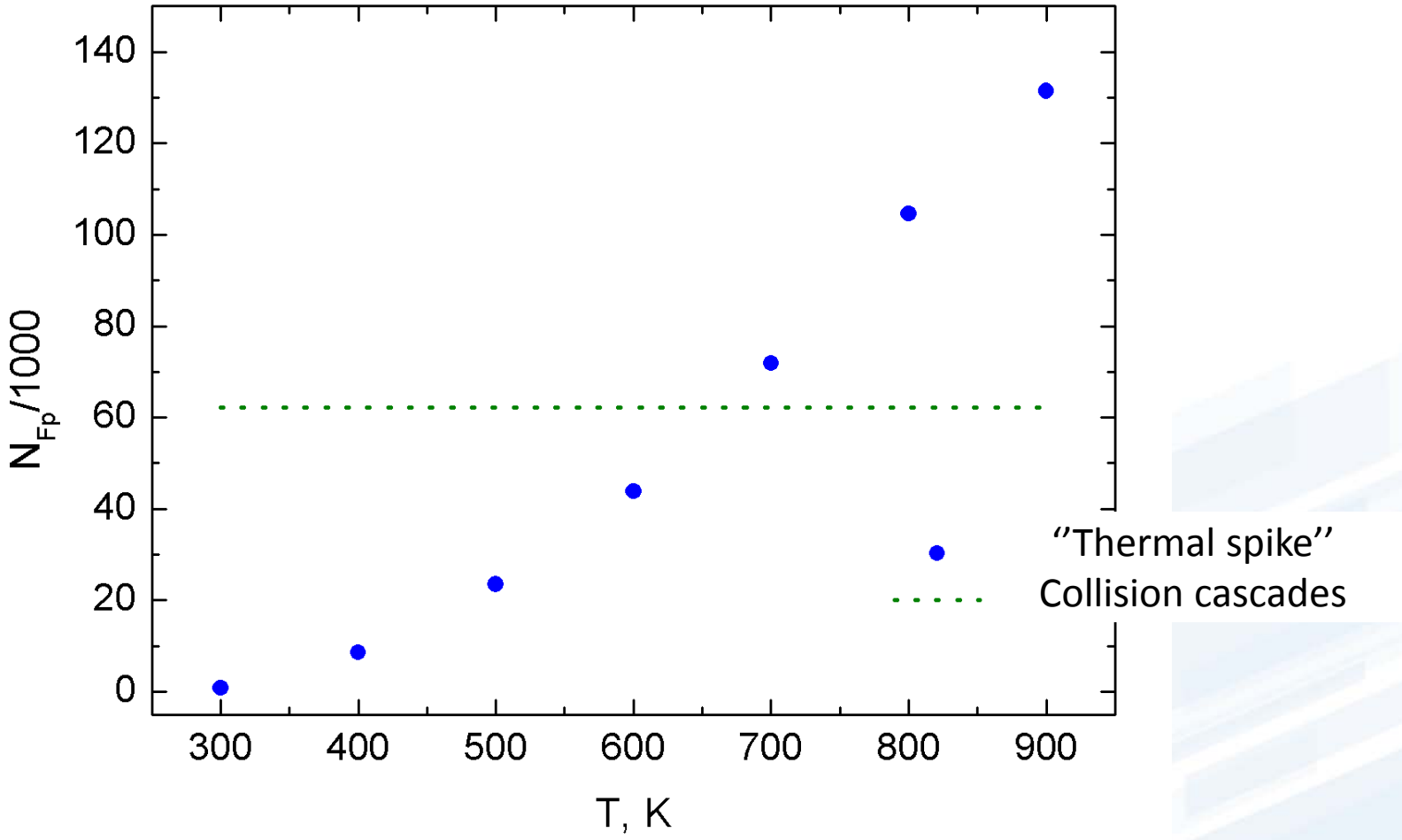
The number of Frenkel pairs dependence on temperature



$N = N_0(S - S_{th})$

$N_0 \approx 2$ keV^{-1}

The number of Frenkel pairs dependence on temperature



TTM parameters

Test

Main

$$C_e(T_e)\rho_e\frac{\partial T_e}{\partial t} = \nabla(\kappa_e \nabla T_e) - g_p(T_e - T_i)$$

g_p = 4,5 10^{17} Вм$^{-1}$К$^{-1}$

g_p = 6,5 10^{17} Вм$^{-1}$К$^{-1}$

ρ_e = 0,625 eÅ^{-3}

$$C_e = \gamma T_e$$

γ = 1,37 10^{-9} эВК$^{-2}$e^{-1}

γ = 4,0 10^{-9} эВК$^{-2}$e^{-1}

$$\kappa_e = D_e \rho_e C_e$$

D_e = 25 Å^2 фс$^{-1}$

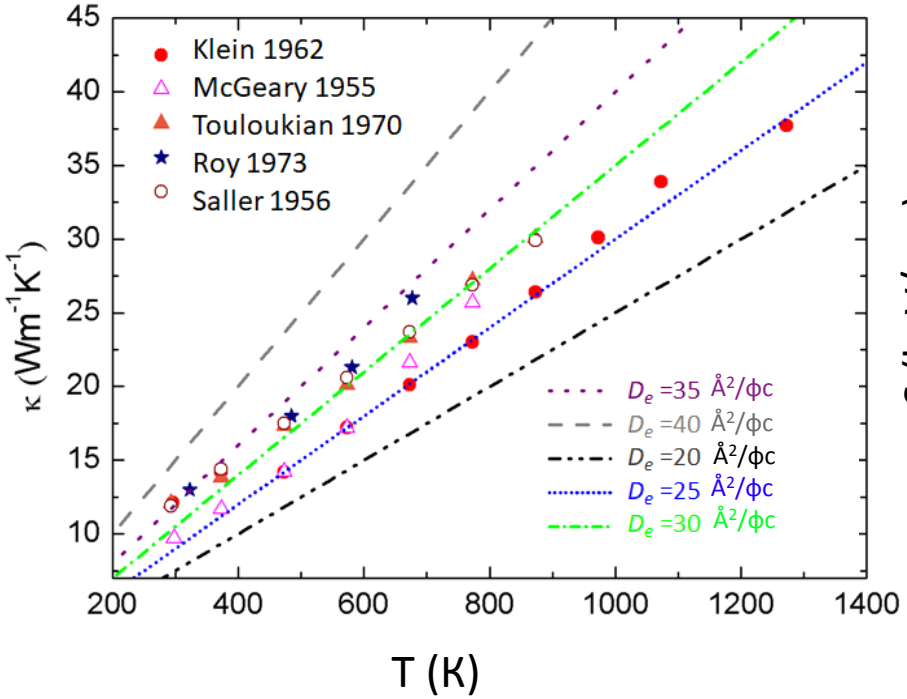
D_e = 10 Å^2 фс$^{-1}$

$$\vec{F}_j = -\nabla_j U(r_1, \ldots r_n) + F_j^{lang}(T_e - T_i)$$

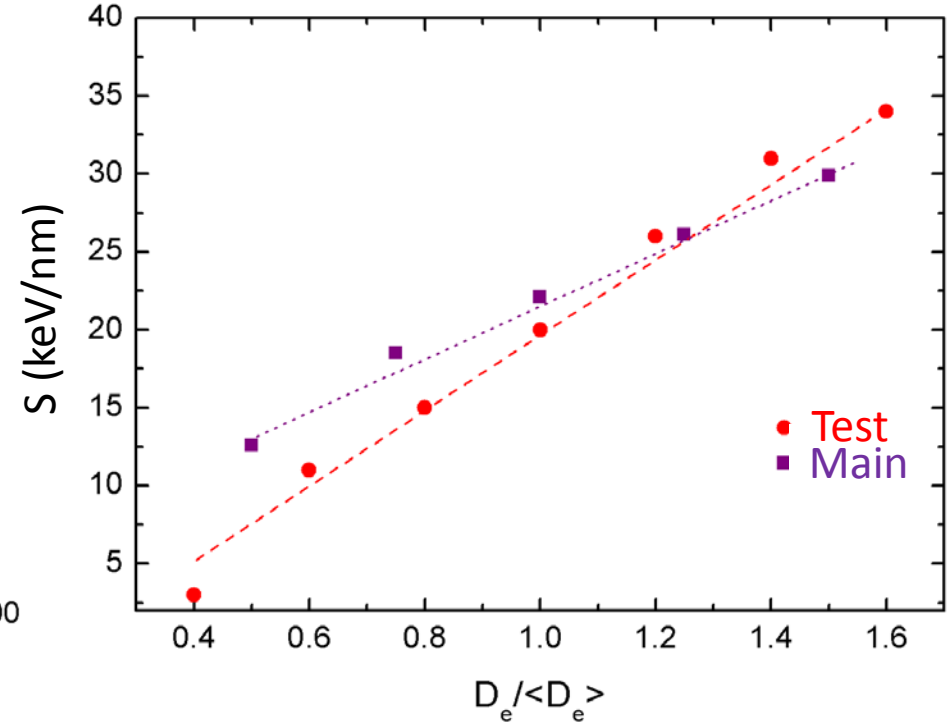
Ni and W
Lin Z, Zhigilei L V et al 2008
J. Phys. Rev. B **77** 075133

Z. Li, C. Wang et al 2017
J. Phys. Plasmas **24** 022703

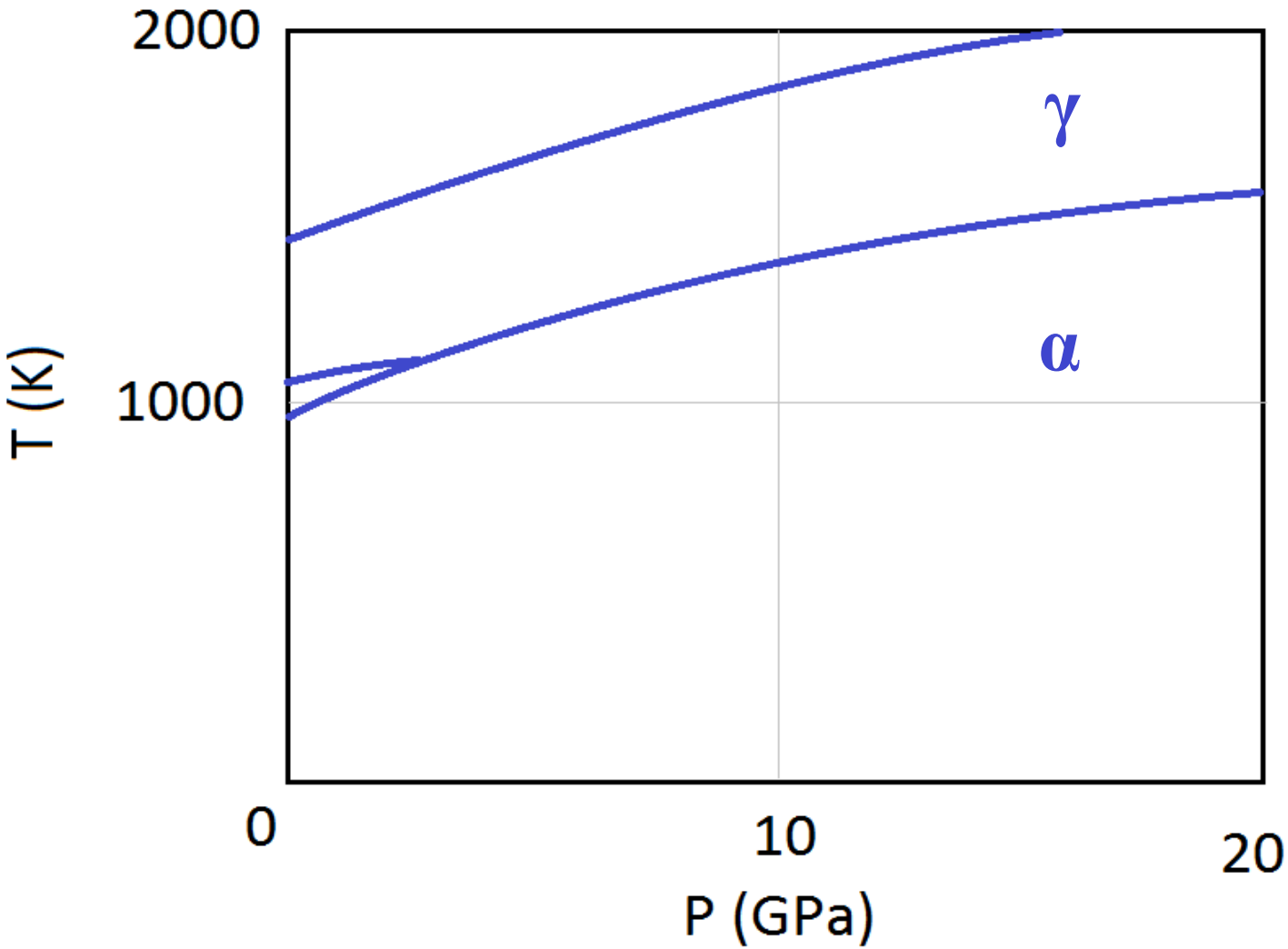
TTM parameters



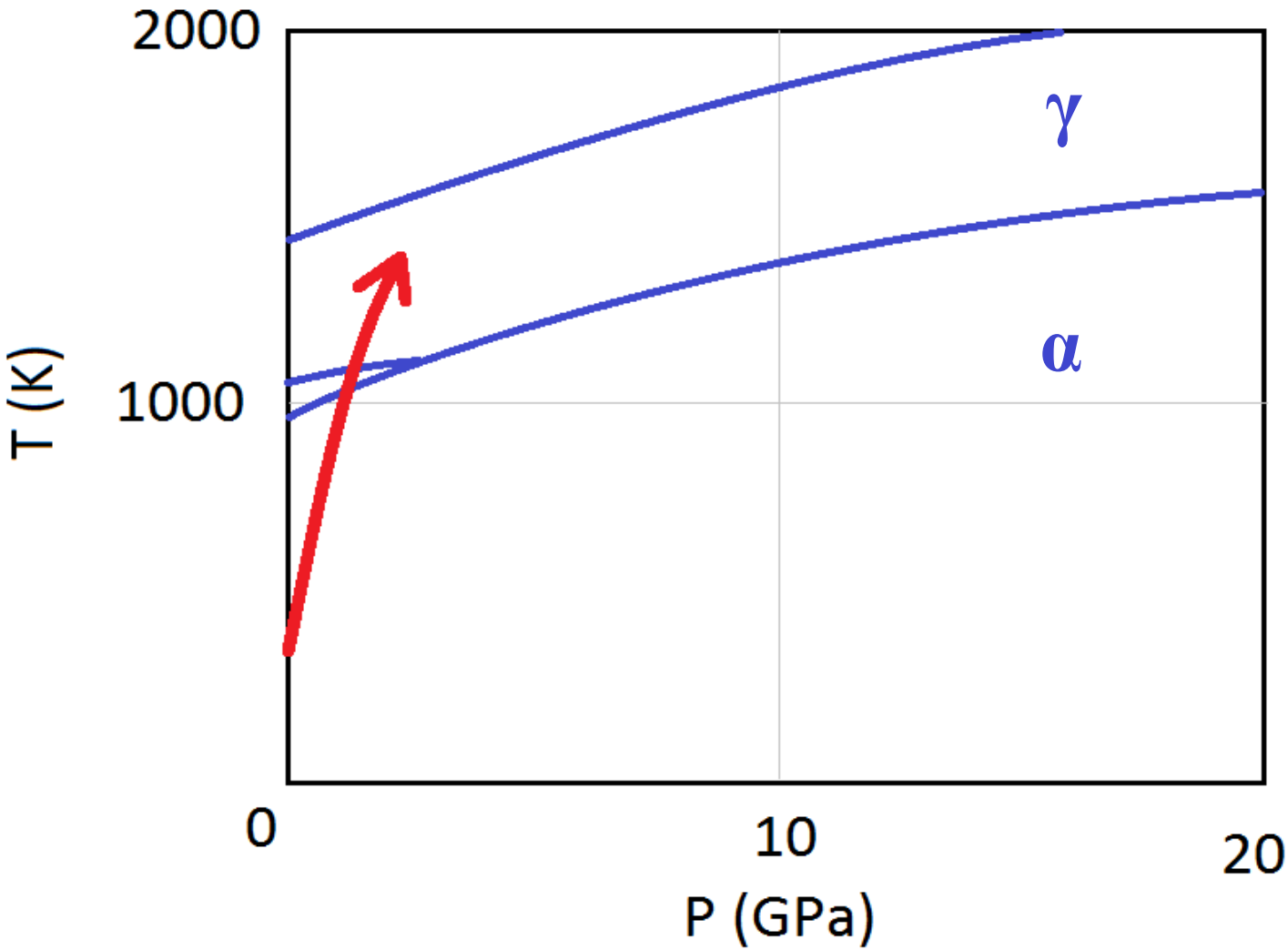
The threshold stopping power



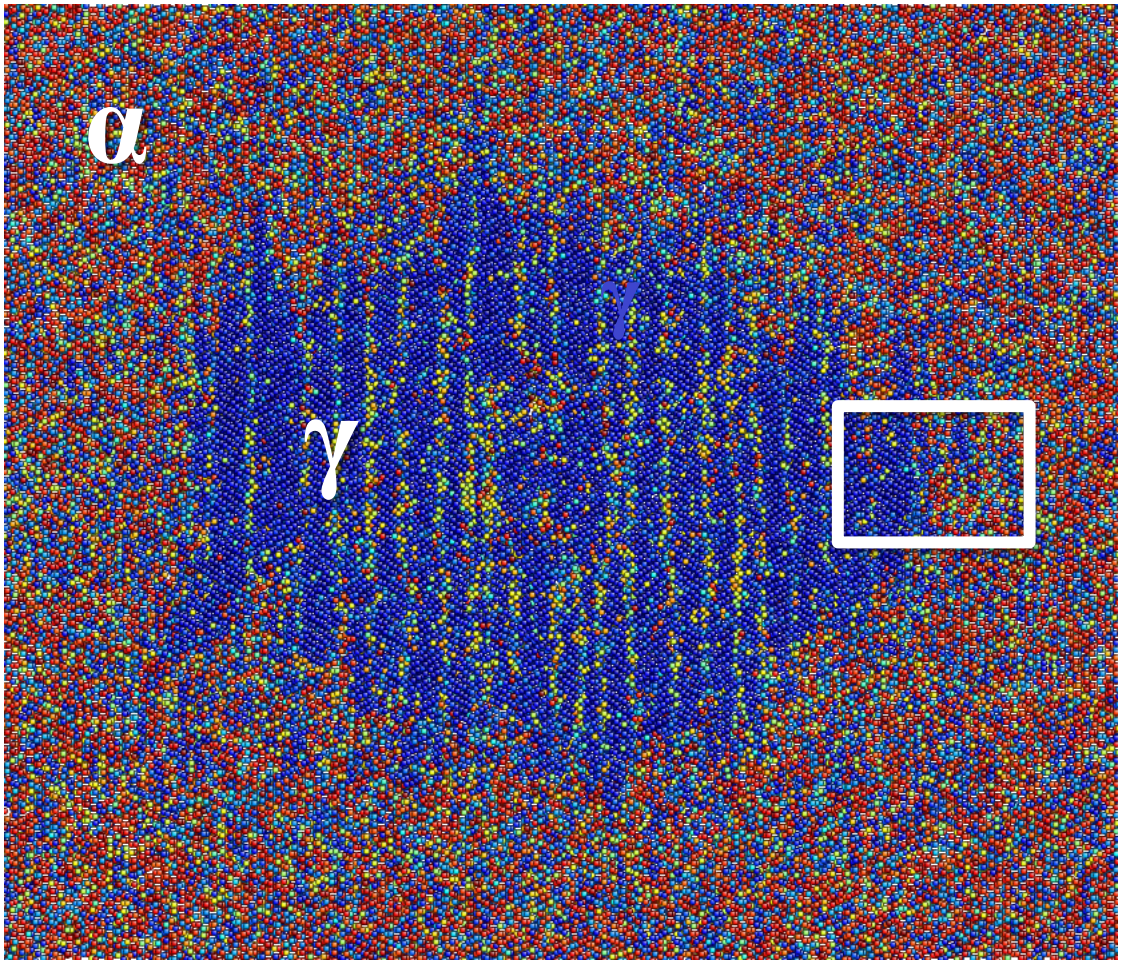
The track formation in α-phase



The track formation in α-phase



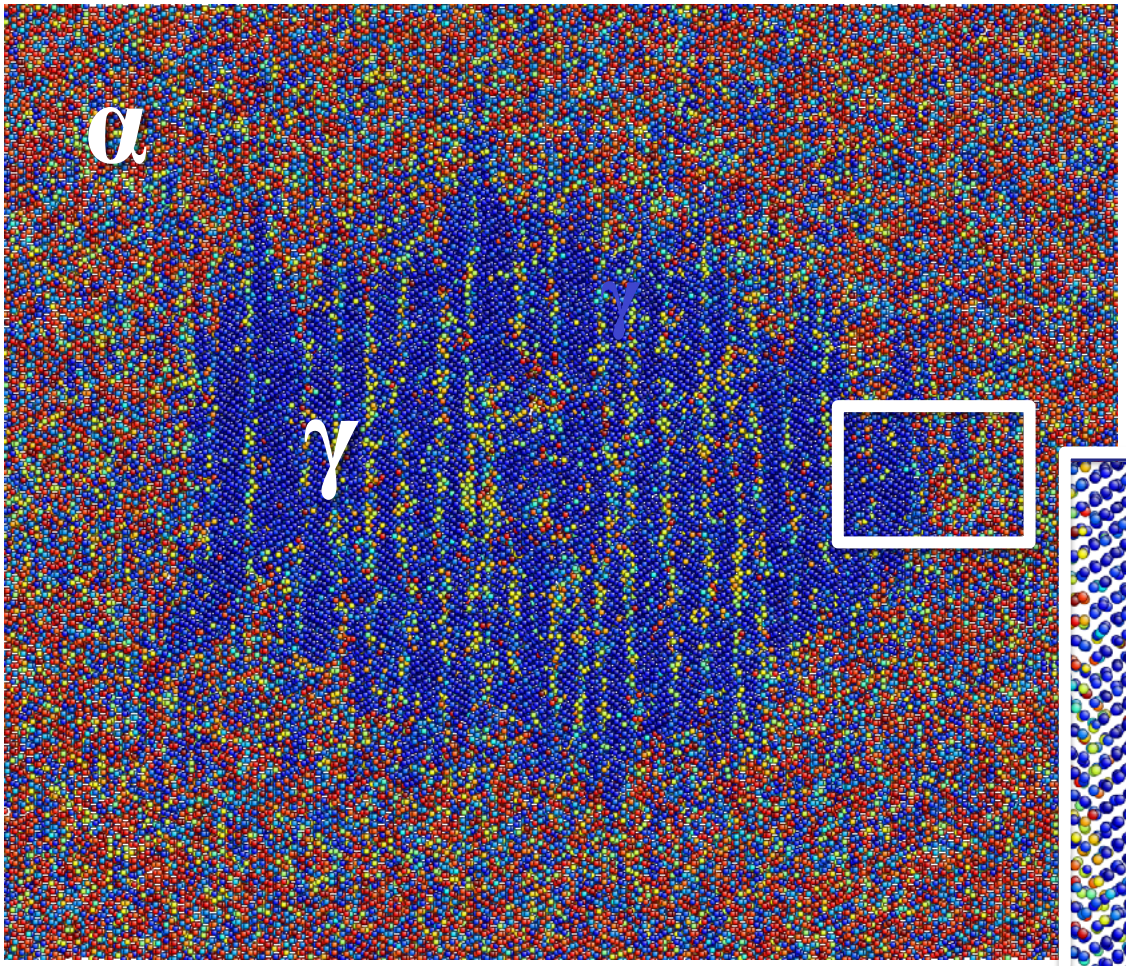
The track formation in α-phase



Threshold stopping power

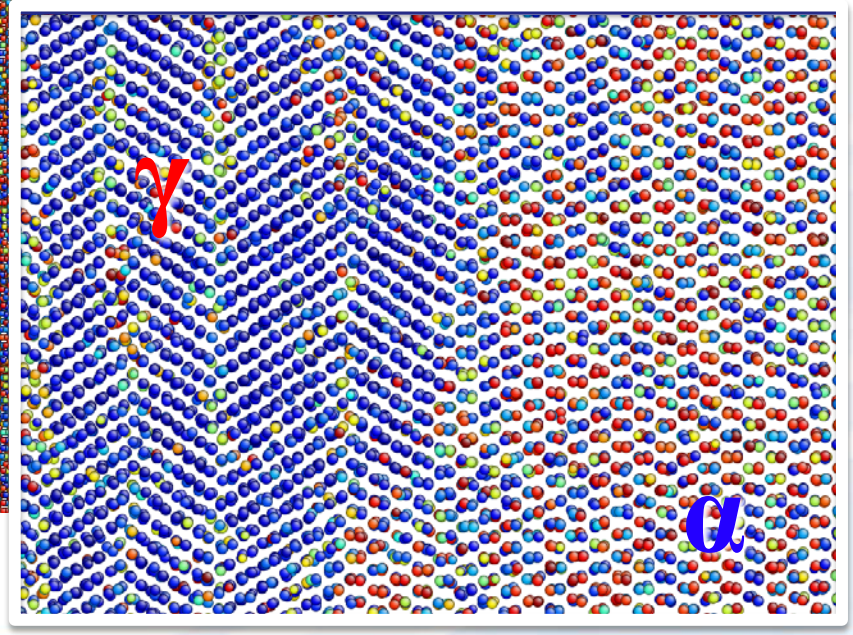
dE/dz = 16 keV/nm

The track formation in α-phase



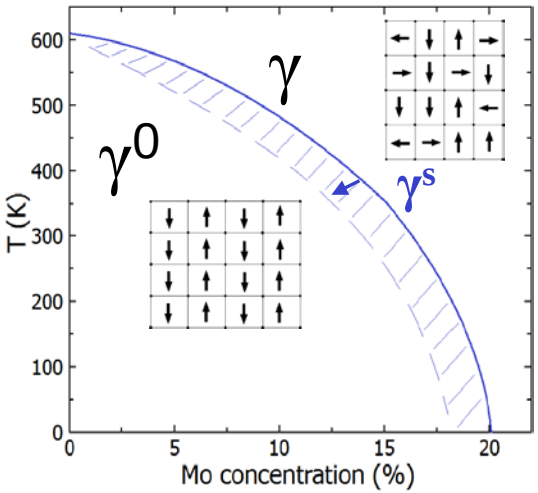
Threshold stopping power

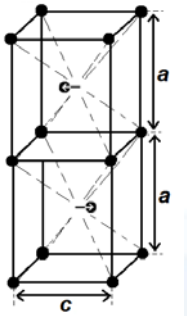
dE/dz = 16 keV/nm

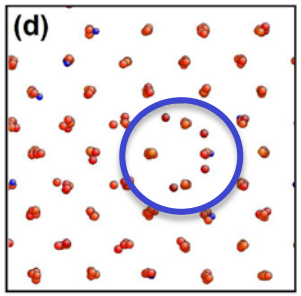


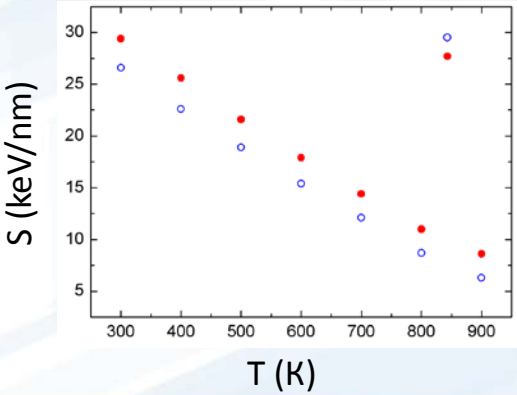
Conclusions

- ✓ The structure of high-temperature γ-phase locally reflects features of γ^0 –phase
- ✓ The martensitic transformation $\gamma \rightarrow \gamma^0$ is quite similar to an atoms rearrangement at ferro- to paraelastic transition of order-disorder type
- ✓ The simulation results indicate that the defects formation in U-Mo may be produced without melting and subsequent crystallization
- ✓ The number of primary radiation defects formed at stage of thermal spike is comparable to the number of primary radiation defects formed in collision cascades, but the number of such defects is weakly dependent on temperature

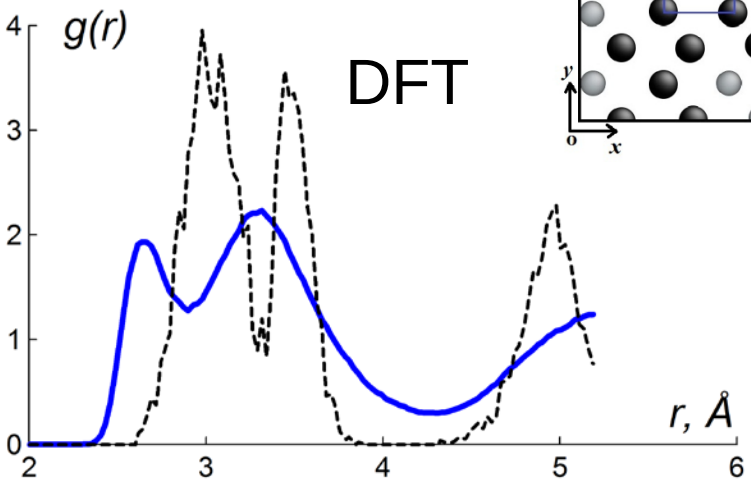


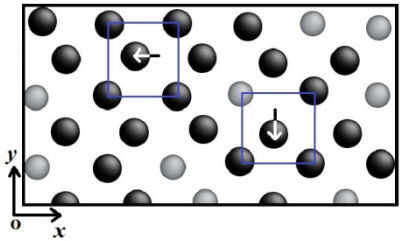


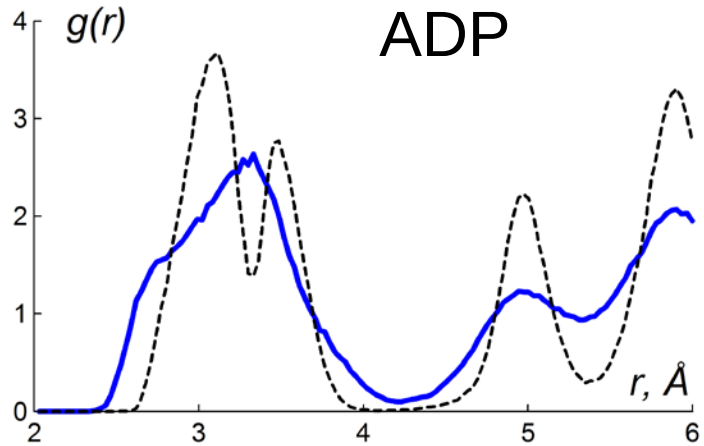


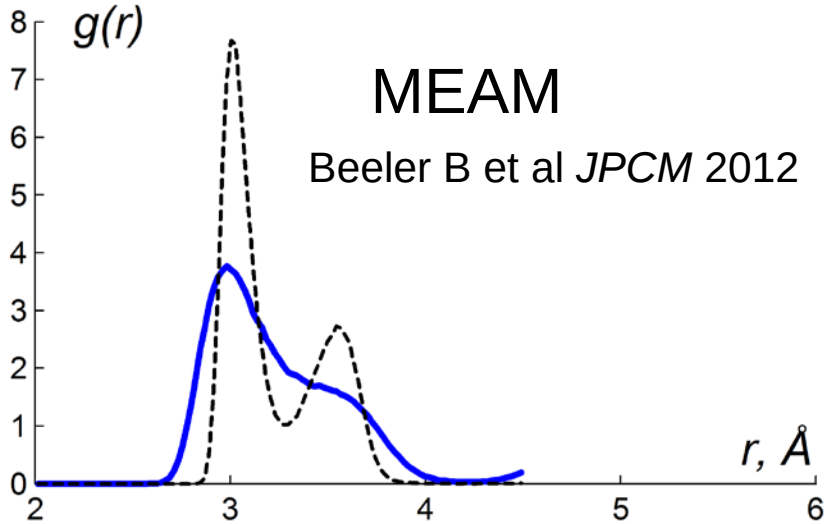


Radial distribution functions of γ-U (T=1000K)





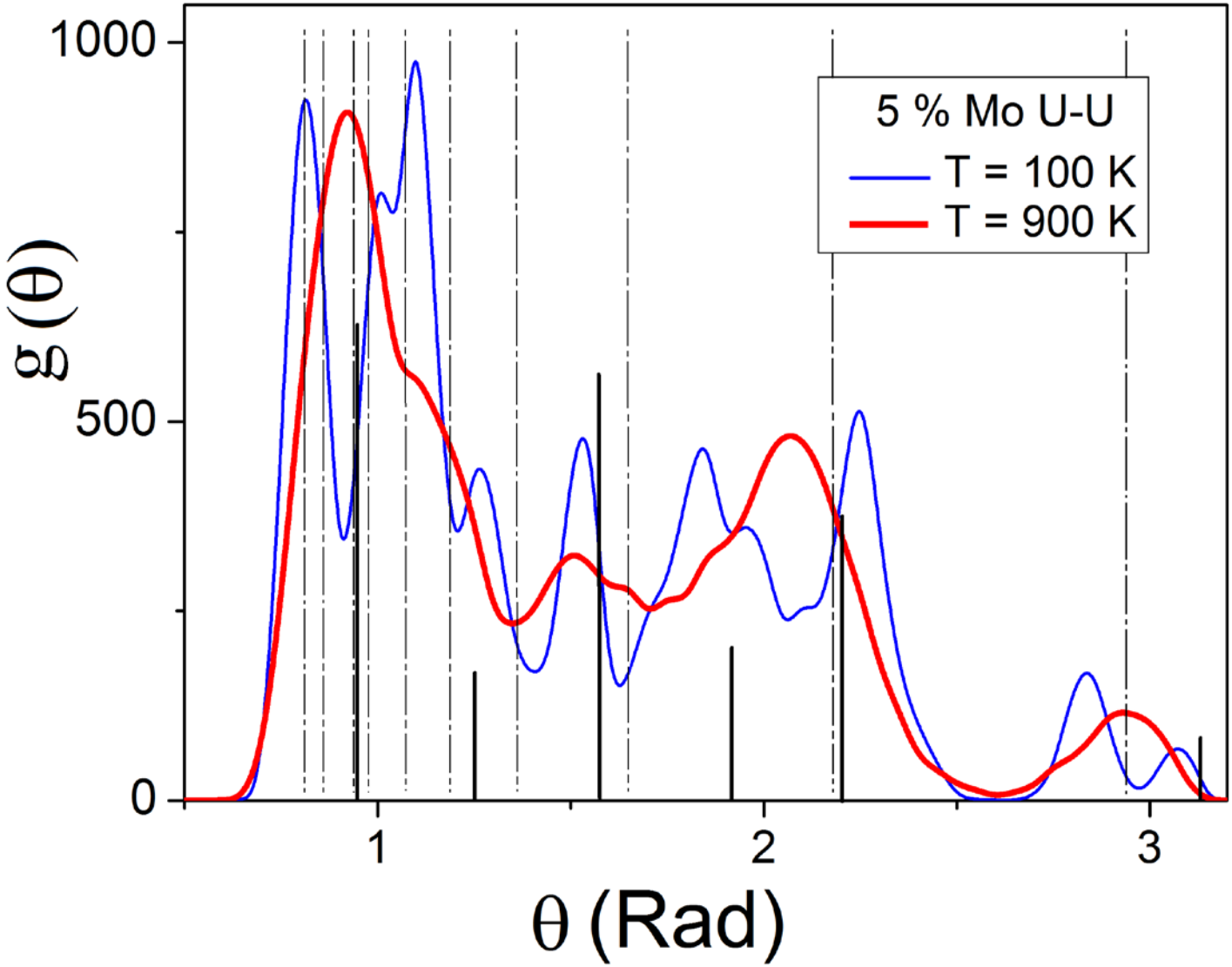




Beeler B et al *JPCM* 2012

RDF G(r)

Averaged RDF G(<r>)

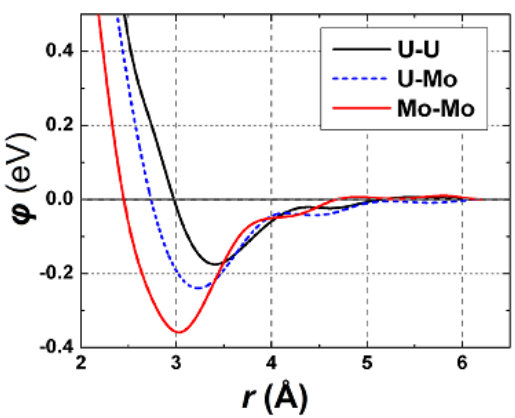

5 % Mo U-U
T = 100 K
T = 900 K
1000
500
0
g(θ)
1
2
3
θ (Rad)

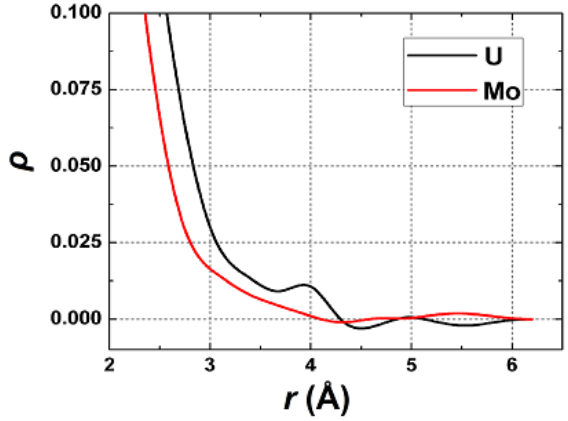
Потенциал для описания системы U-Mo:

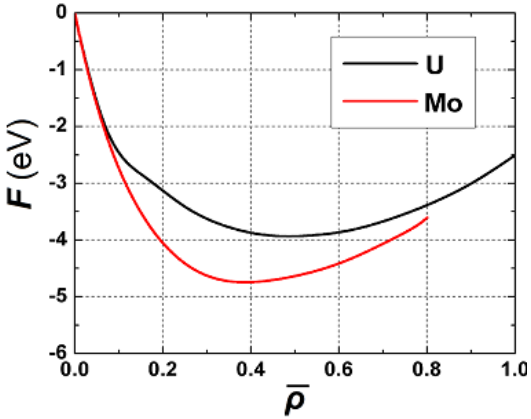
ADP — Angular-dependent potential:

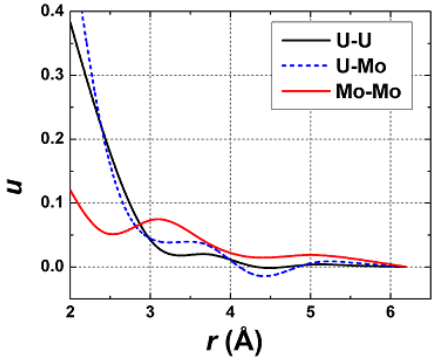
$$E_i = F_\alpha\left(\sum_{j\neq i}\rho_\beta(r_{ij})\right) + \frac{1}{2}\sum_{j\neq i}\phi_{\alpha\beta}(r_{ij}) + \frac{1}{2}\sum_{s}(\mu_i^s)^2 + \frac{1}{2}\sum_{s,t}(\lambda_i^{st})^2 - \frac{1}{6}\nu_i^2$$

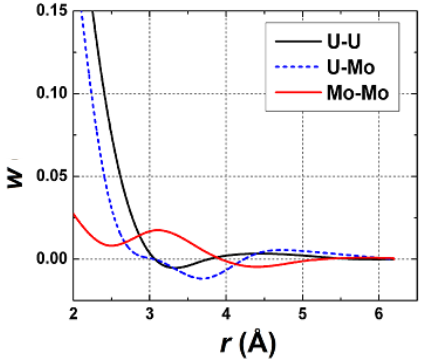
***ADP*: Y. Mishin, Mehl *et al* 2005 *Acta Mater* 53 4029**











$$\mu_i^s = \sum_{j\neq i} u_{\alpha\beta}(r_{ij}) r_{ij}^s$$

$$\lambda_i^{st} = \sum_{j\neq i} w_{\alpha\beta}(r_{ij}) r_{ij}^s r_{ij}^t$$

$$\nu_i = \sum_s \lambda_i^{ss}$$

Потенциал для описания системы U-Mo:

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***ADP*: Y. Mishin, Mehl *et al* 2005 *Acta Mater* 53 4029**

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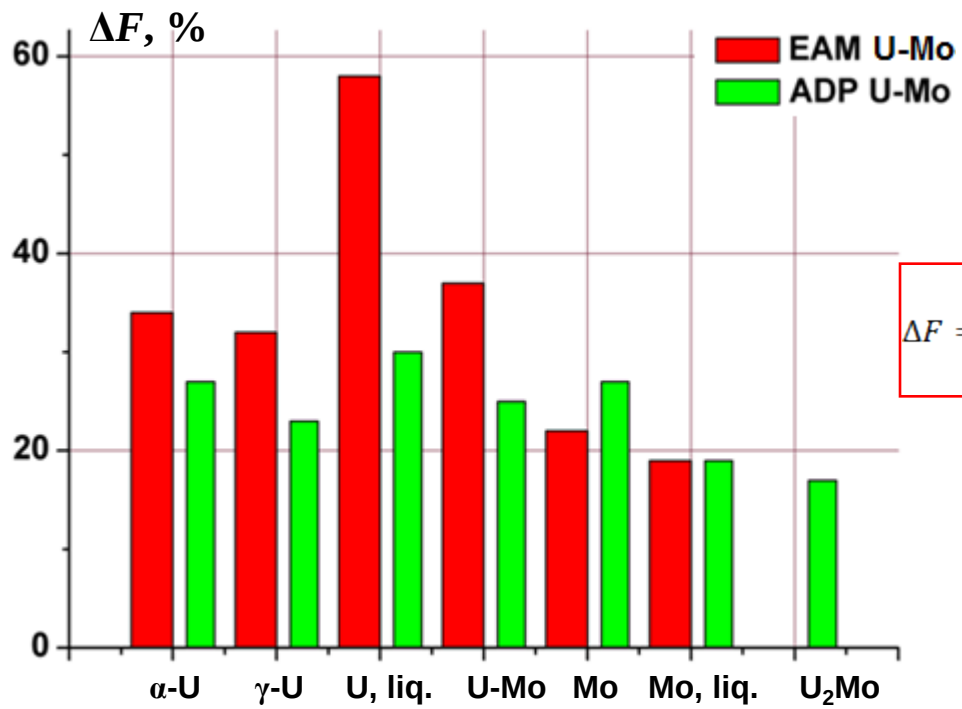
$$\lambda_i^{st} = \sum_{j\neq i} w_{\alpha\beta}(r_{ij}) r_{ij}^s r_{ij}^t$$

$$\nu_i = \sum_s \lambda_i^{ss}$$

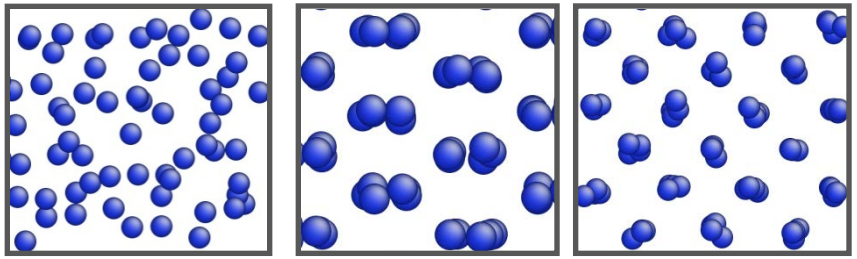


$$\Delta F = \frac{1}{N}\sum_{i=1}^{N}\sqrt{\frac{(F_{ADP}^i - F_{DFT}^i)^2}{(F_{DFT}^i)^2}} \times 100, \%$$

EAM: Smirnova D. E., Kuksin A. Y., Starikov S. V. et al. // MSM Sci. Eng. 2013. V. 21. P. 035011.

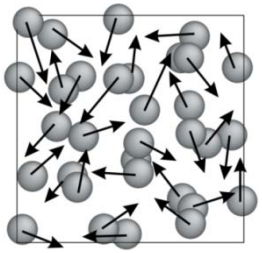
ADP: Smirnova D. E., Kuksin A. Y., Starikov S. V. //JNM. 2015. V. 458. Pp. 304-311.

Метод «согласования по силе»:



Расплав U **α–U** **γ–U**

VASP



$$\mathbf{F}_i = -\frac{\partial E}{\partial \mathbf{R}_i}$$

$$E = \frac{\langle \varphi_n | \mathbf{H} | \varphi_n \rangle}{\langle \varphi_n | \varphi_n \rangle}$$

F, E, S

potfit

$$Z_F = \sum_{i=1}^{N_\alpha} \sum_{\alpha=x,y,z} \sqrt{\frac{(F^i_{ADP} - F^i_{DFT})^2}{(F^i_{DFT})^2}}$$

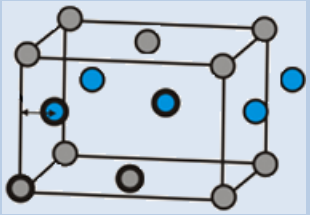
***Потенциал межатомного взаимодействия* (ADP):**

$$E_i = F_\alpha\left(\sum_{j\neq i} \rho_\beta(r_{ij})\right) + \frac{1}{2}\sum_{j\neq i} \phi_{\alpha\beta}(r_{ij}) + \frac{1}{2}\sum_s (\mu_i^s)^2 + \frac{1}{2}\sum_{s,t} (\lambda_i^{st})^2 - \frac{1}{6}\nu_i^2$$

Верификация потенциала путем расчета большого набора характеристик материала и сопоставления результатов с экспериментальными данными

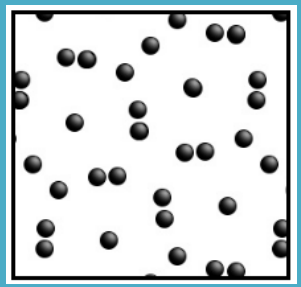
Force-matching: F. Ercolessi, J.B. Adams 1994 *EPL* **26** 583 ***VASP 5.2:*** G. Kresse 1996 *Phys. Rev. B* **54** 11169
Potfit: P. Brommer, F. Gahler F 2006 *Phil. Mag.* **86** 753 ***ADP:*** Y. Mishin, Mehl *et al* 2005 *Acta Mater* **53** 4029

Verification of created ADP potential (pure U)



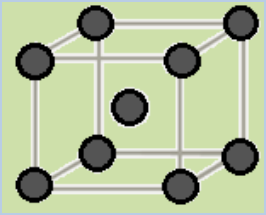
α-U	*a* (Å)	*b* (Å)	*c* (Å)	*C11* (GPa)	*C22* (GPa)	*C33* (GPa)	*C12* (GPa)	*C13* (GPa)	*C23* (GPa)
ADP U-Mo	**2.849**	**5.841**	**4.993**	**253**	**199**	**265**	**93**	**90**	**105**
Experiments*	2.854	5.870	4.955	215	199	267	46	22	108

* C. Barrett et al. Phes. Rev. **129** (1963) 625



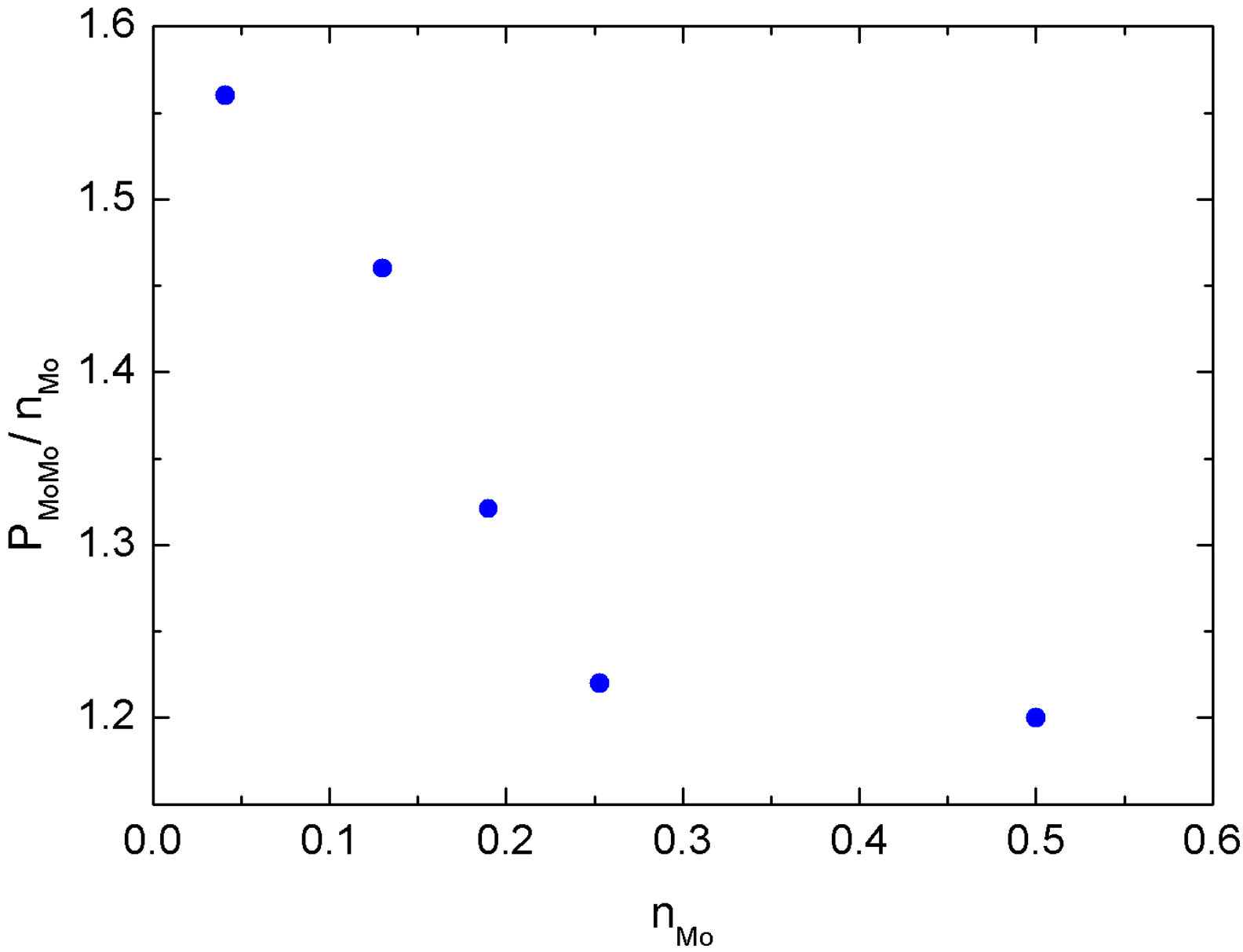
β-U	symmetry	*a* (Å)	*c* (Å)	E_β-E_α (eV/atom)	T_{tr} (K)
ADP U-Mo	**$P4_2nm$**	**10.612**	**5.603**	**0.089**	**780**
Experiments	$P4_2nm$	10.590	5.634	> 0	1045
***Calculations* (*GGA*)**	$P4_2nm$	10.460	5.660	0.099	—

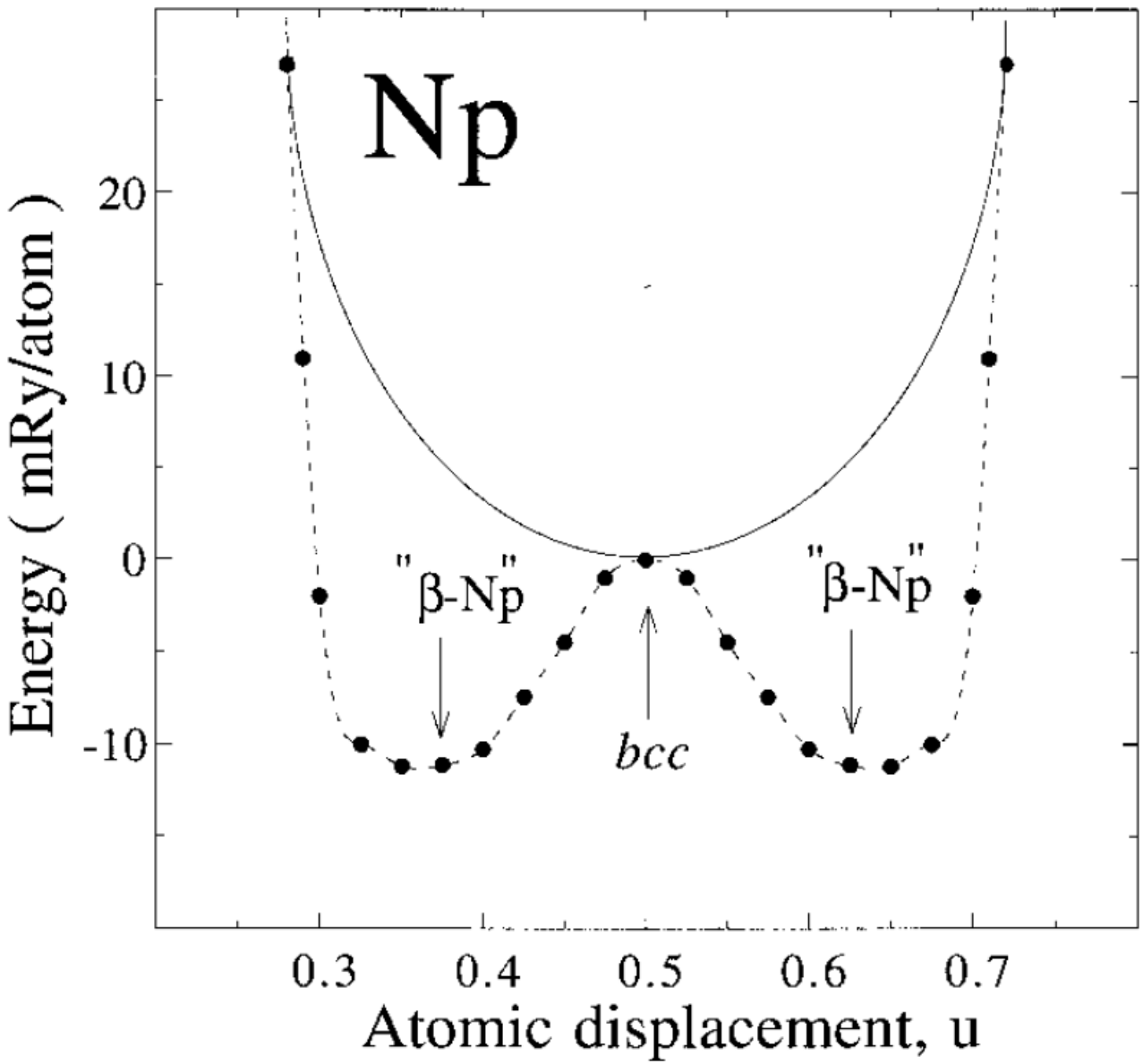
*Experiment: C. Tucker and P. Senio, Acta Crystallographica **6** (1953) 753760* *GGA: Y. Li et al. JNM **475*** (2016)



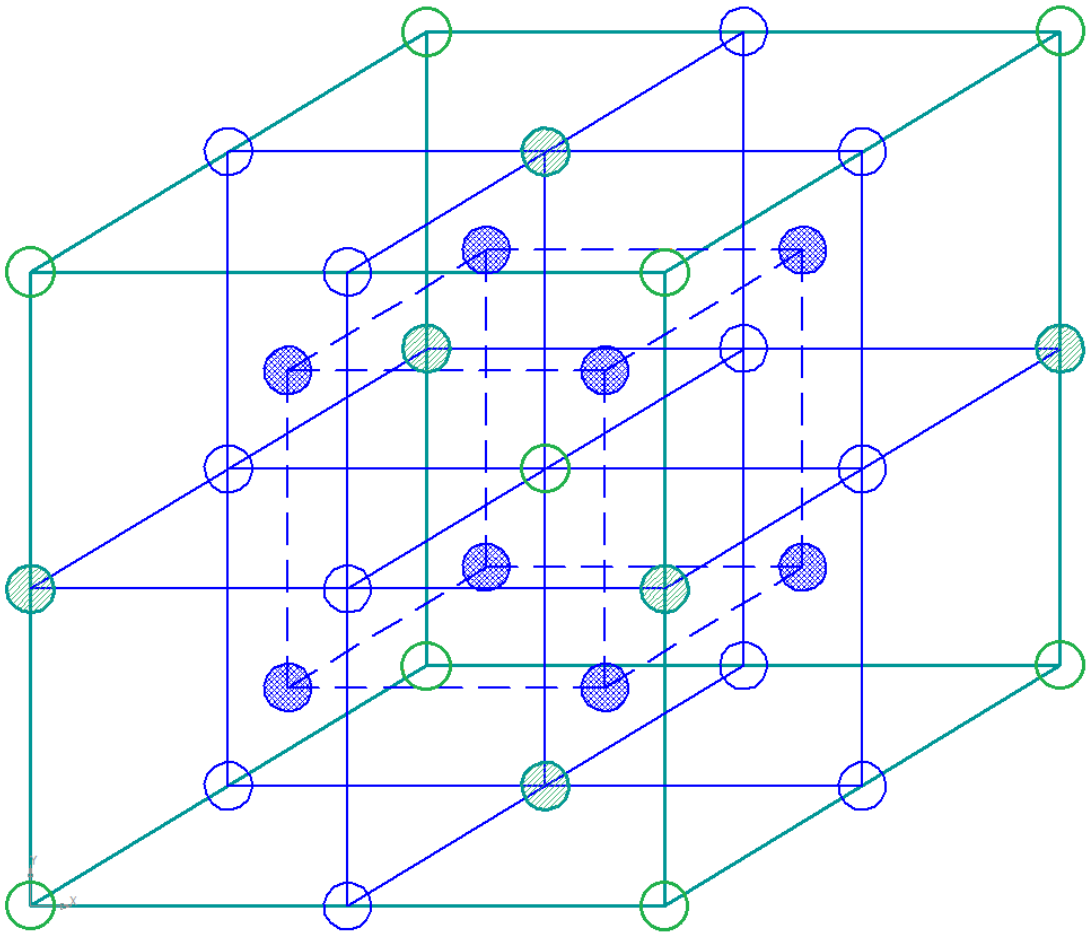
γ-U (T=1000K)	*a* (Å)	B (GPa)	E_{bcc}-E_α (eV/atom)	T_{melt} (K)	E^f_{vac} (eV)
ADP U-Mo	**3.52**	**120**	**0.072**	**1330**	**2.1**
Experiments	3.47	113	> 0	1408	0.9 –1.45
***Calculations* (*GGA*)**	3.43	136	0.28	—	1.2

*Experiment: C.-S. Yoo et al. Phys. Rev. B. **57** (1998) 10359.* *GGA: Y. Li et al. JNM **475*** (2016)


1.6
1.5
1.4
1.3
1.2
P_{MoMo} / n_{Mo}
0.0
0.1
0.2
0.3
0.4
0.5
0.6
n_{Mo}


Np
Energy (mRy/atom)
20
10
0
-10
"β-Np"
"β-Np"
bcc
0.3
0.4
0.5
0.6
0.7
Atomic displacement, u

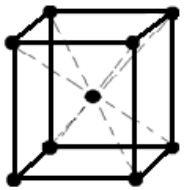
γ^S structure



Energy hierarchy of phases in pure uranium

$E - E_{\alpha}$ (eV/atom)

bcc

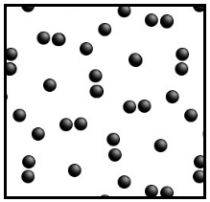


0.24 - 0.28 — **DFT [Y. Li et al. JNM 475 (2016)]**
[C. Taylor PRB 77 (2008)]

≈

0.15 — **DFT [R.Q. Hood et al. PRB 78 (2008)]**
[J.P. Crocombette et al. PRB 64 (2001)]

β-U

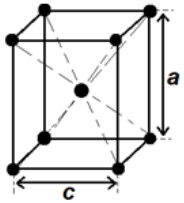


0.1 — **DFT [Y. Li et al. JNM 475 (2016)]**

β 0.89 — ADP potential (this work)

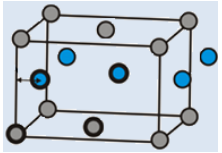
bcc 0.72 — ADP potential (this work)

bct



bct 0.05 — ADP potential (this work)

0.04 — **DFT [R.Q. Hood et al. PRB (2008)]**



0.02 — DFT & ADP potential (this work)

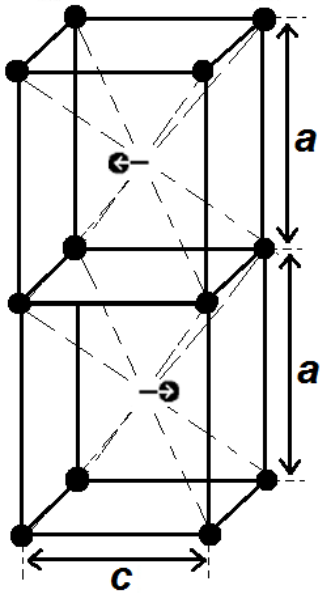
0 — **α-U**

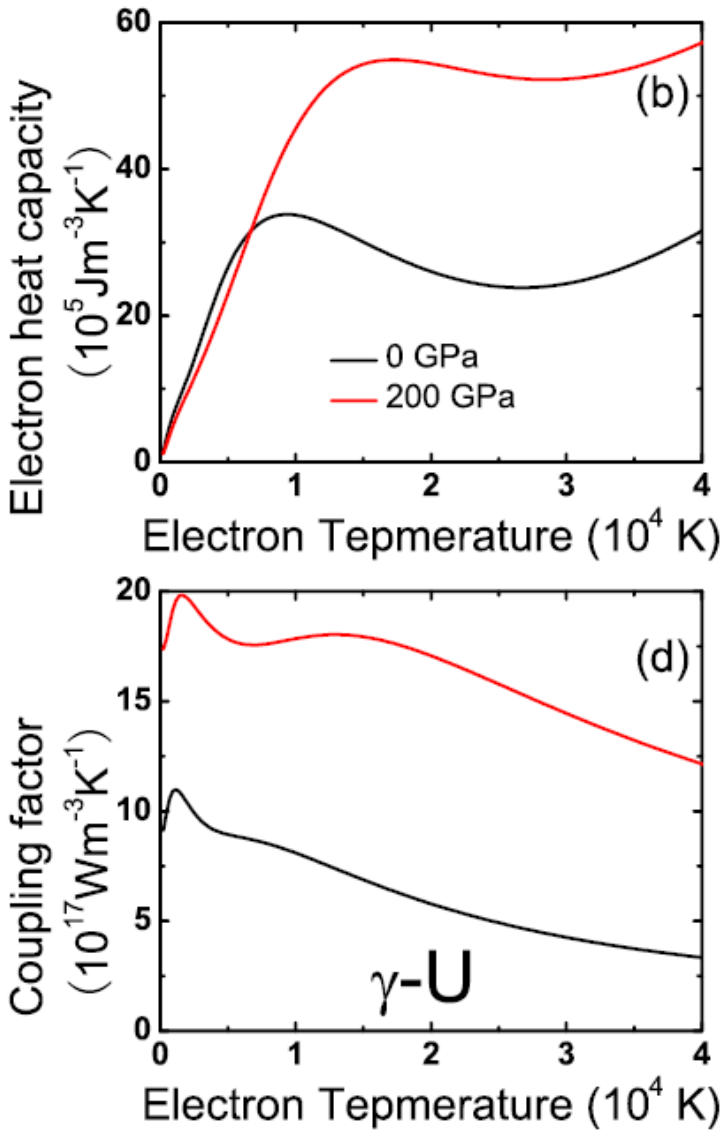
Experimentally observed phase for the alloys

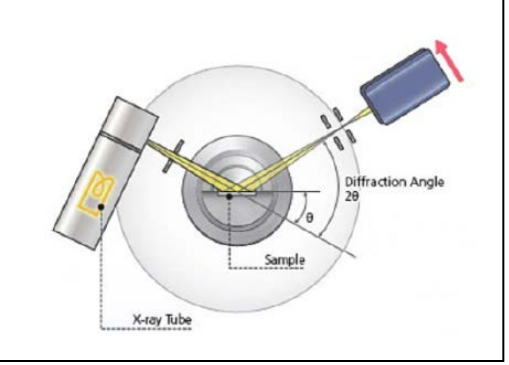
[K. Tangri and G.I. Williams, JNM **4** (1961)]
[B.W. Howlett , JNM **35** (1970)]
[H.L. Yakel, JNM **33** (1969)]
[N.T. Chebotarev et al. Atomnaya Energiya **48** (1980)]

$c < a = b$

γ^0




(b)
Electron heat capacity
(10^5Jm^{-3}K^{-1})
0 GPa
200 GPa
Electron Tepmerature (10^4 K)
(d)
Coupling factor
(10^{17}Wm^{-3}K^{-1})
γ-U
Electron Tepmerature (10^4 K)



1) В обратном пространстве задается сетка расчетных точек.

2) Интенсивности рассеяния рассчитываются в узлах сетки. Если расчетная точка не совпала с узлом обратной решетки (где интенсивность велика), то весомого вклада в пункте 3 она не даст.

3) Происходит суммирование по узлам с равными углами рассеяния.

4) 2-3 повторяются на заданных шагах интегрирования, происходит усреднение.

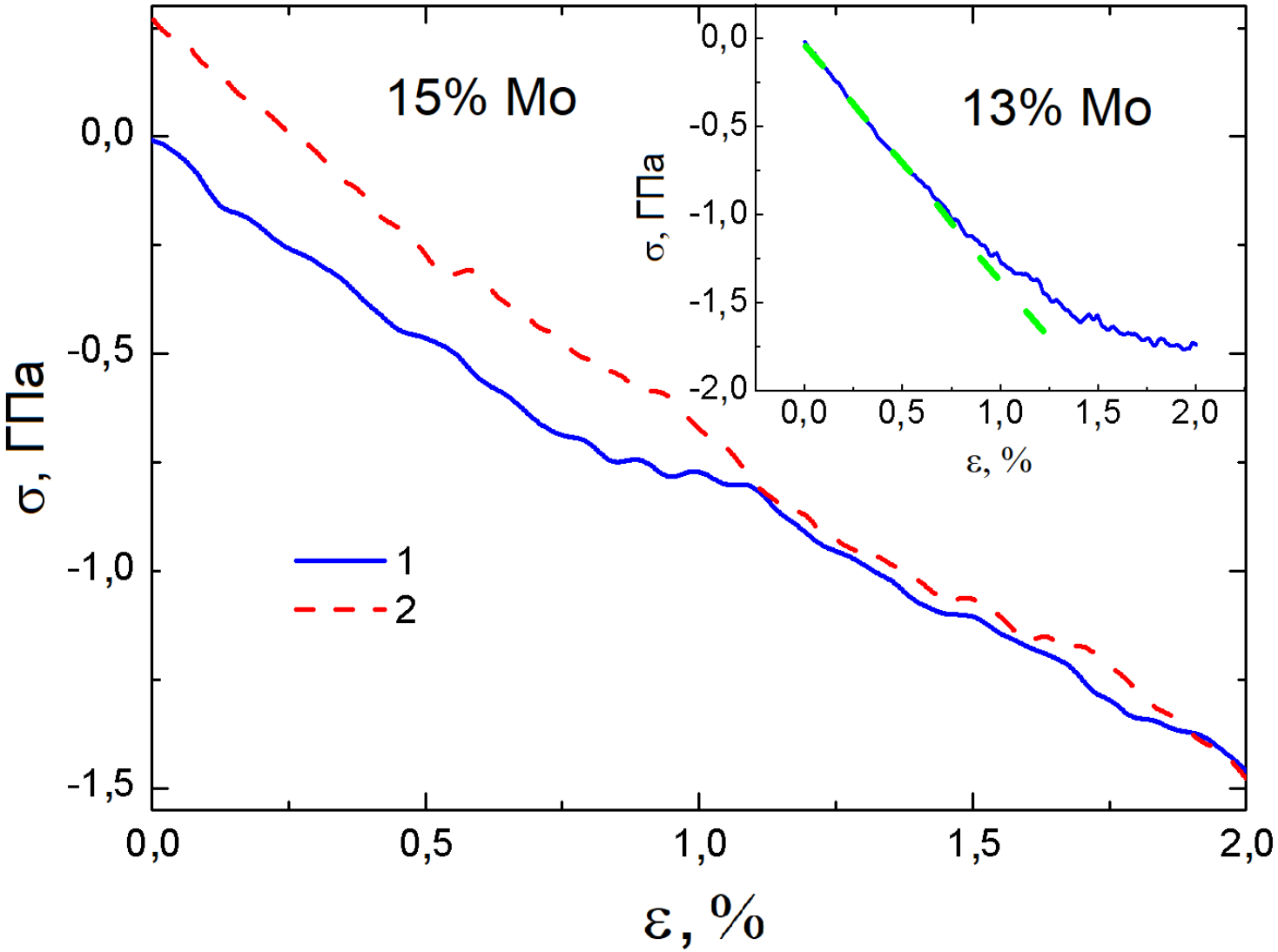
$$\mathbf{A} = 2\pi\frac{[b,c]}{(a,b,c)}, \mathbf{B} = 2\pi\frac{[c,a]}{(a,b,c)}, \mathbf{C} = 2\pi\frac{[a,b]}{(a,b,c)}, \qquad \mathbf{K}_{hkl} = h\mathbf{A} + k\mathbf{B} + l\mathbf{C}$$

$$\mathbf{k} - \mathbf{k}' = \mathbf{K}_{hkl}$$

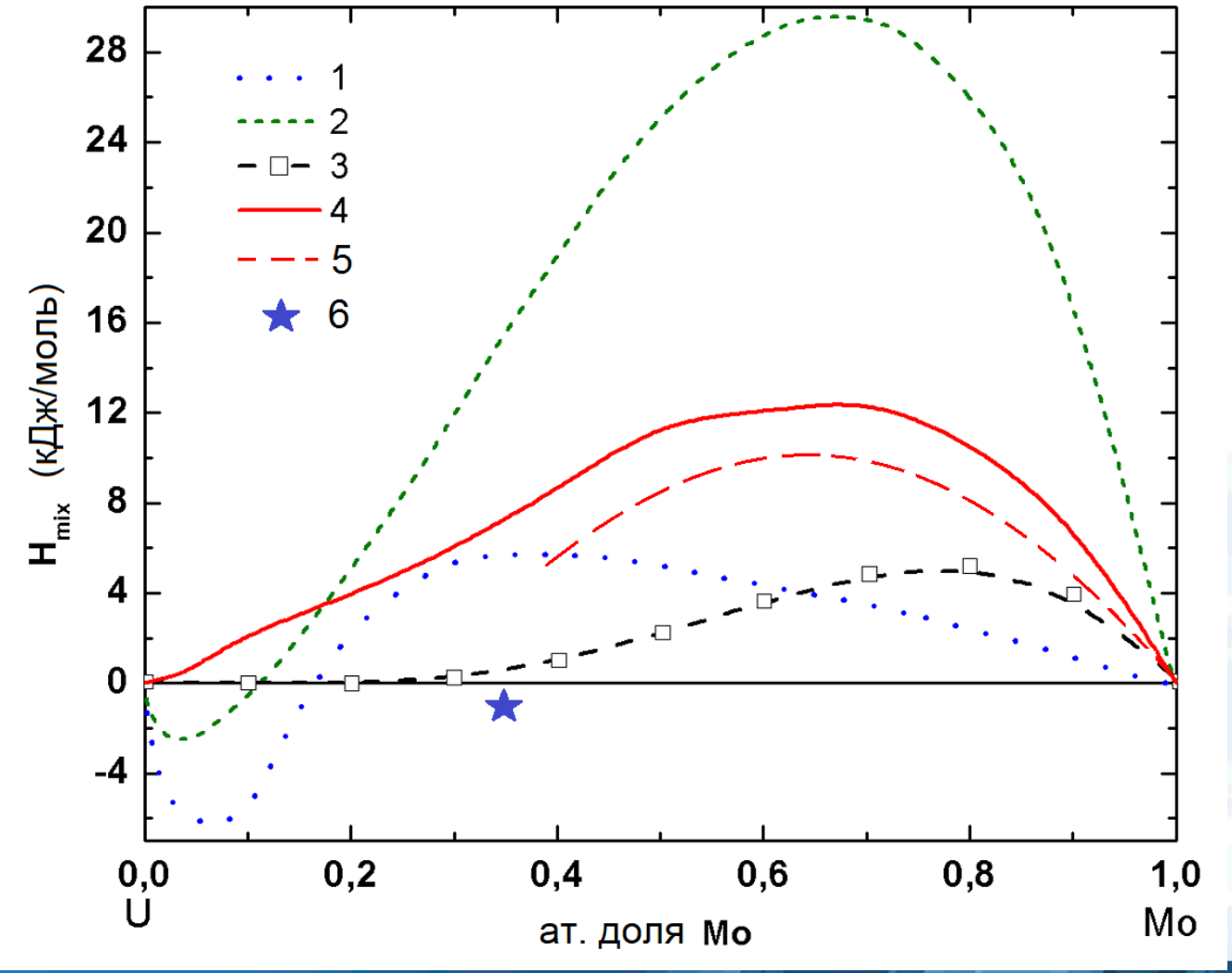
$$I = P(\theta)\, F F^*$$

$$F = \sum_{j=1} f_j(\theta) \exp\{-2i\pi \mathbf{r}_j \cdot \mathbf{K}_{hkl}\} = \sum_{j=1} f_j(\theta) \exp\{-2i\pi(x_j h + y_j k + z_j l)\},$$

$$\mathbf{r}_j = x_j\mathbf{a} + y_j\mathbf{b} + z_j\mathbf{c} \qquad \sin(\theta) = \frac{\lambda|\mathbf{K}|}{2} = \frac{\lambda}{2d_{hkl}},$$

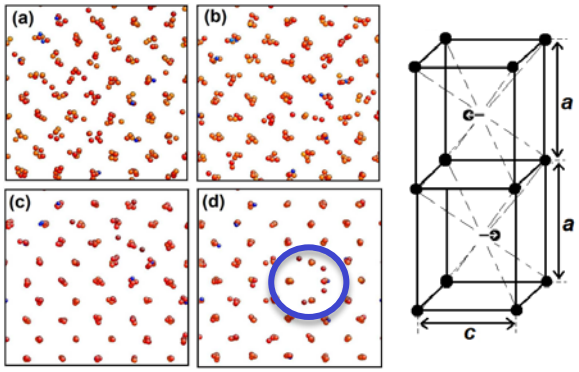

15% Mo
13% Mo
σ, ГПа
ε, %
1
2
0,0
-0,5
-1,0
-1,5
-2,0
0,5
1,0
1,5
2,0

Enthalpy of mixing of U-Mo alloy

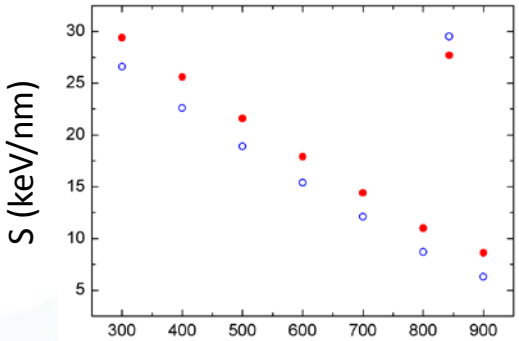


Conclusions

- ✓ The simulation results indicate that the defects formation in U-Mo may be produced without melting and subsequent crystallization



- ✓ The threshold stopping power of swift ions dependence on temperature was obtained



- ✓ The number of primary radiation defects formed at stage of thermal spike is comparable to the number of primary radiation defects formed in collision cascades, but the number of such defects is weakly dependent on temperature