РОСАТОМ

ГОСУДАРСТВЕННАЯ КОРПОРАЦИЯ ПО АТОМНОЙ ЭНЕРГИИ «РОСАТОМ»

# Prospects for plasma de-excitation of $^{186m}$Re nuclear isomers

**Vladimir Koltsov**

**Khlopin Radium Institute**

**Saint-Petersburg**

***E-mail: vladimir-koltsov@yandex.ru***

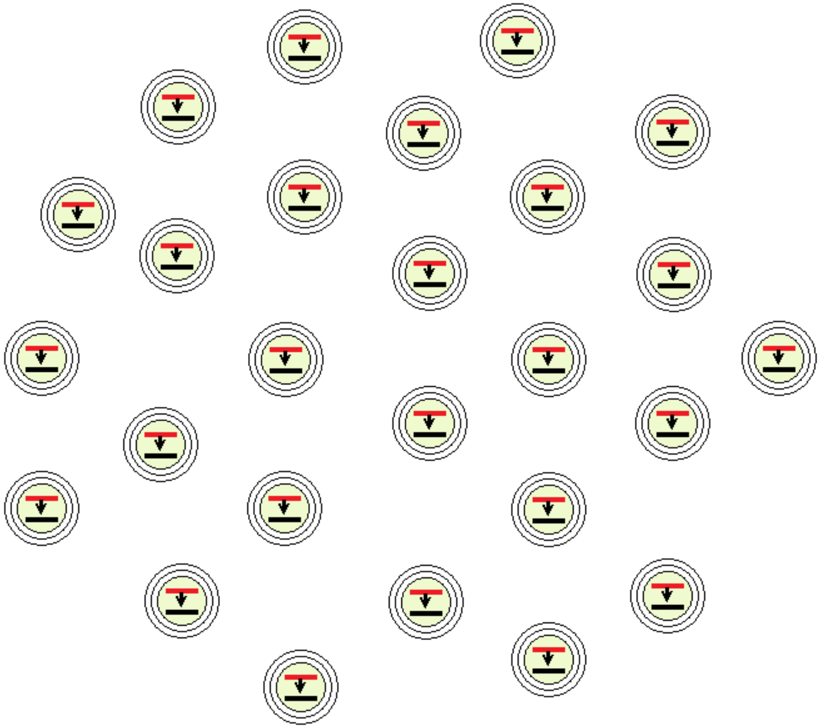
# Nuclear isomers in Khlopin Radium Institute

1. Study of the influence of the medium on the probability of spontaneous isomeric transitions in nuclei.
2. Participation in study on stimulated de-excitation of nuclear isomers in laser plasma.
3. Development of radiochemical technologies for the production and purification of nuclear isomeric material.
4. Manufacturing of targets and sources with nuclear isomers.

Nuclear isomerism was discovered in the 1930s
by I.V. Kurchatov with collaborators from Ioffe Physical-Technical Institute
and by L.V. Mysovsky with collaborators from Khlopin Radium Institute.

# The overall goal of the research

The overall is the creation of a controlled source of nuclear energy based on the stimulation of the de-excitation of nuclear isomers.



The problem is to stimulate mass de-excitation of nuclear isomers in an avalanche-like or external action with high efficiency.

Up to now this problem has not been solved.

# The need for isomeric energy sources

- Power sources with a power of ~ 1 μW have been developed ($^{63}Ni$) .
- The question of powerful sources remains open: low specific power, high environmental hazard, inconvenience of operation.
- Solution is power source via stimulating de-excitation of the nuclear isomer $^{186m}Re$.

| Isotope | Half life | Starting isotope | σ (n, γ), barn | Energy capacity, J/g | Power , W/g | |
|---|---|---|---|---|---|---|
| | | | | | working | storage |
| $^{63}Ni$ | 100 y | $^{62}Ni$ nat. 4 % | 15 | $3 \cdot 10^{7}$ | $6 \cdot 10^{-3}$ | |
| $^{238}Pu$ | 87.7 y | $^{237}Np$ reactor | 169 | $2 \cdot 10^{9}$ | $6 \cdot 10^{-1}$ | |
| $^{210}Po$ | 138 d | $^{209}Bi$ nat. 100 % | 0.03 | $2 \cdot 10^{9}$ | $1.4 \cdot 10^{2}$ | |
| $^{186m}Re$ isomer | 200 000 y | $^{185}Re$ nat. 37 % | 0.3 | $1 \cdot 10^{8}$ | $3 \cdot 10^{2}$ | $3 \cdot 10^{-5}$ |

# Outline

1. Introduction

− Spontaneous nuclear isomeric transitions.

− Possible ways to stimulate the de-excitation of nuclear isomers.

− Review of non-plasma studies with isomeric nuclei.

− Review of studies with isomeric nuclei in laser plasma.

Conclusion: an isomeric energy source has failed, but a lot of material has been accumulated.

2. Prospects for the study of the $^{186m}$Re isomer in an electro-discharge plasma.

3. Possibility of creating an energy source based on the $^{186m}$Re isomer.

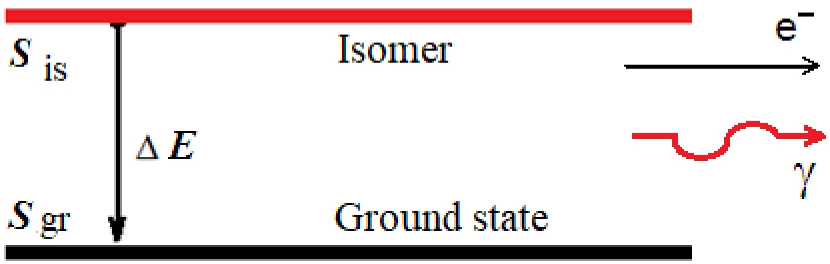
The main goal:

Show the possibility of currently creating a controlled energy source

based on stimulated de-excitation of the $^{186m}$Re isomer.

# Spontaneous isomeric transitions

A long-lived isomer is formed when there is a large difference in the structure of nuclear states, for example, in the spins $S_{is}$ and $S_{gr}$.



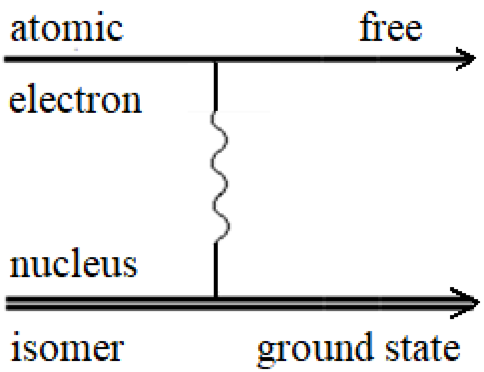
Nuclear energy levels

1. The probability of emission of $\gamma$-quantum with a wavelength $\lambda$

$$P_\gamma \sim \alpha \frac{c}{\lambda} \left( \frac{R_{nucl.}}{\lambda} \right)^{2L}$$

$$\alpha = \frac{e^2}{\hbar c} \approx \frac{1}{137}, \quad L = \left| S_{is} - S_{gr} \right|$$

2. The probability of conversion transition




$$P_{conv.} = \beta_{conv.} \, P_\gamma$$

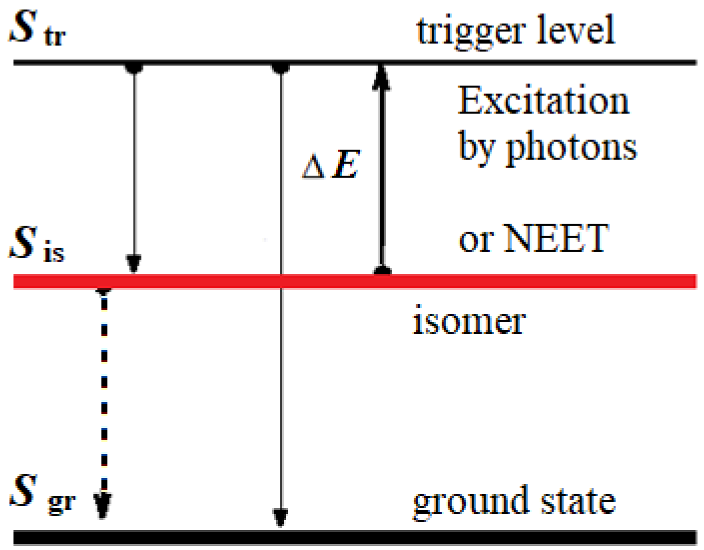
$$\beta_{conv.} \sim \alpha \, (\alpha Z)^3 \left( \frac{2 m_e c^2}{\Delta E} \right)^{L+5/2} >> 1$$ for $\Delta E < 10$ keV

For isomeric transitions of low energy, electron conversion is more likely, and vice versa – the excitation of the isomer is more efficient via the excitation of the electron shell of the atom.

# Methods for stimulating the de-excitation of the isomeric state

De-excitation of the isomer via the trigger level →

Direct stimulation of the isomeric transition by resonant photon radiation →




$$S_{gr} < S_{tr} < S_{is}$$

1. Irradiation of the isomer with photons at the transition frequency $\omega \approx \Delta E / \hbar$ :

– stimulation of direct isomeric transition (the idea of a gamma laser has not yet worked out);

– excitation of the trigger level.

2. Excitation of the trigger level via the energy transfer from the atomic shell to the nucleus (NEET).

The multipolarity of the trigger transition is less than the multipolarity of the direct isomeric transition.

For known trigger transitions $\Delta E \geq 1$ keV.

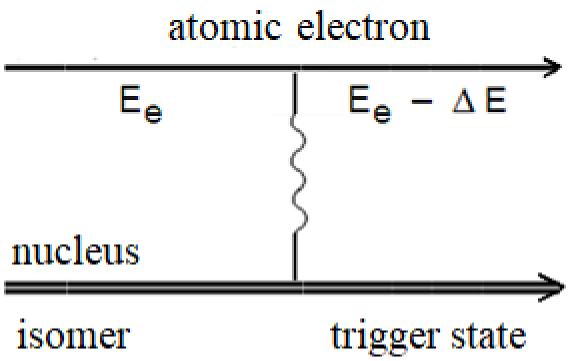
# **Methods for trigger level excitation** (*reviews: Tkalya, 2004; Karamian , 2008*)

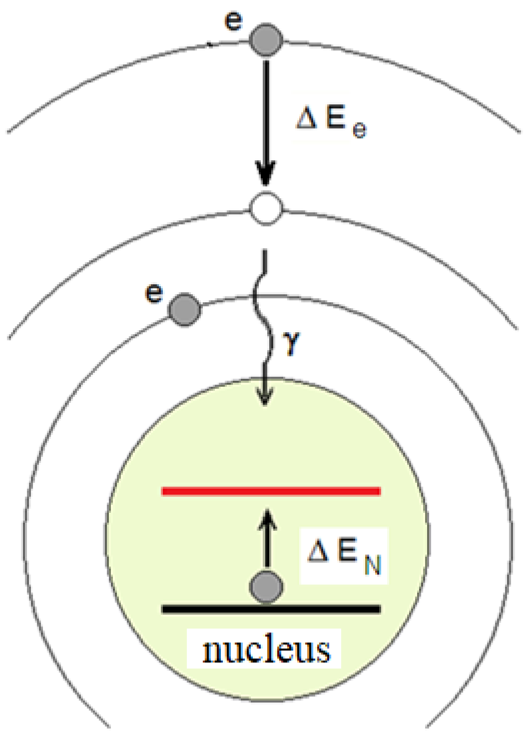
1. Excitation of the isomeric nucleus by photons $\hbar\omega = \Delta E_N$ (for plasma radiation *Haihgt & Baldwin, 1985*).

2. Inelastic scattering of electrons by an isomeric nucleus (for the 235mU isomer: *Grechukhin and Soldatov, 1976*).

3. NEET is the transfer of excitation to the isomeric nucleus during electronic transition between atomic levels (Morita, 1973). We need a resonance between atomic and nuclear transitions.

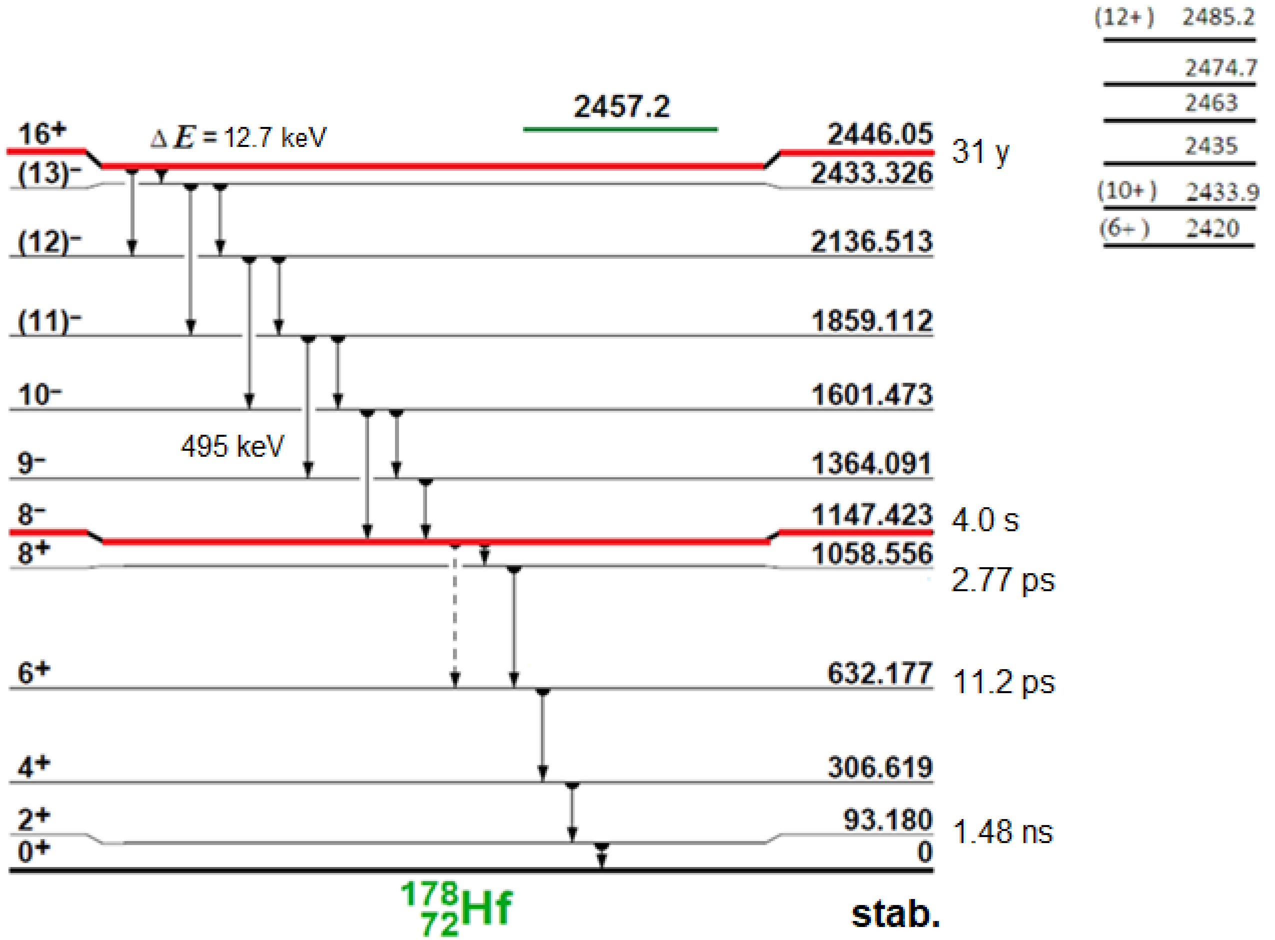
4. Resonant NEET - to achieve resonance, irradiation of an excited atom with an isomeric nucleus by photons $\hbar\omega = \Delta E_e - \Delta E_N$ (*Zon and Karpeshin, 1990*).




NEET scheme. $\Delta E_e \approx \Delta E_N$

# Isomer $^{178m2}$Hf - an interesting subject of research *( Collins,... 1999)*




- Mainly targets with the $^{178m2}$Hf isomer were irradiated with X-rays.
- There is no unambiguous experiment to stimulate the de-excitation of the isomer.
- Feature - for the preparation of targets by radiochemistry, it is difficult to separate the ground and isomeric states of $^{178}$Hf nuclei.

## **Promising isomers $T_{1/2}$ > 3 days** (*Karamian , 2008* + recent data )

| Isomer | $T_{1/2}$ | $E_{is}$, keV |
|---|---|---|
| $^{91m}$Nb | 61 d | 105 |
| $^{92m}$Nb | 16,1 y | 31 |
| $^{97m}$Tc | 90 d | 97 |
| $^{102m}$Rh | 2,9 y | 141 |
| $^{108m}$Ag | 418 y | 109 |
| $^{110m}$Ag** | 250 d | 118 |
| $^{113m}$Cd | 14,1 y | 264 |
| $^{114m1}$In | 49,5 d | 180 |
| $^{117m}$Sn | 13,6 d | 315 |
| $^{119m}$Sn * | 293 d | 90 |
| $^{121m}$Sn | 55 y | 6,3 |
| $^{121m}$Te | 154 d | 294 |
| $^{123m}$Te * | 119,7 d | 248 |
| $^{125m}$Te * | 57,4 d | 145 |
| $^{127m}$Te | 109 d | 88 |
| $^{129m}$Te | 33,6 d | 106 |

| Isomer | $T_{1/2}$ | $E_{is}$, keV |
|---|---|---|
| $^{129m}$Xe | 8,9 d | 236 |
| $^{131m}$Xe | 11,8 d | 164 |
| $^{148m}$Pm | 41,3 d | 138 |
| $^{166m}$Ho | 1200 d | 6 |
| $^{174m}$Lu | 142 d | 171 |
| $^{177m}$Lu * | 161 d | 970 |
| $^{178m2}$Hf * | 31 y | 2446 |
| $^{179m2}$Hf | 25 d | 1106 |
| $^{180m}$Ta | $>10^{15}$ y | 75 |
| $^{184m}$Re | 169 d | 188 |
| $^{186m}$Re ** | $2 \cdot 10^{5}$ y | 149 |
| $^{192m}$Ir | 241 y | 155 |
| $^{193m}$Ir | 10,5 y | 80 |
| $^{193m}$Pt | 4,33 d | 150 |
| $^{195m}$Pt | 4,02 d | 259 |
| $^{242m}$Am | 141 y | 49 |

*   de-excitation of isomers was announced without the use of plasma, there is no confirmation,

**  plasma experiments were carried out, positive effect at $^{186m}$Re.

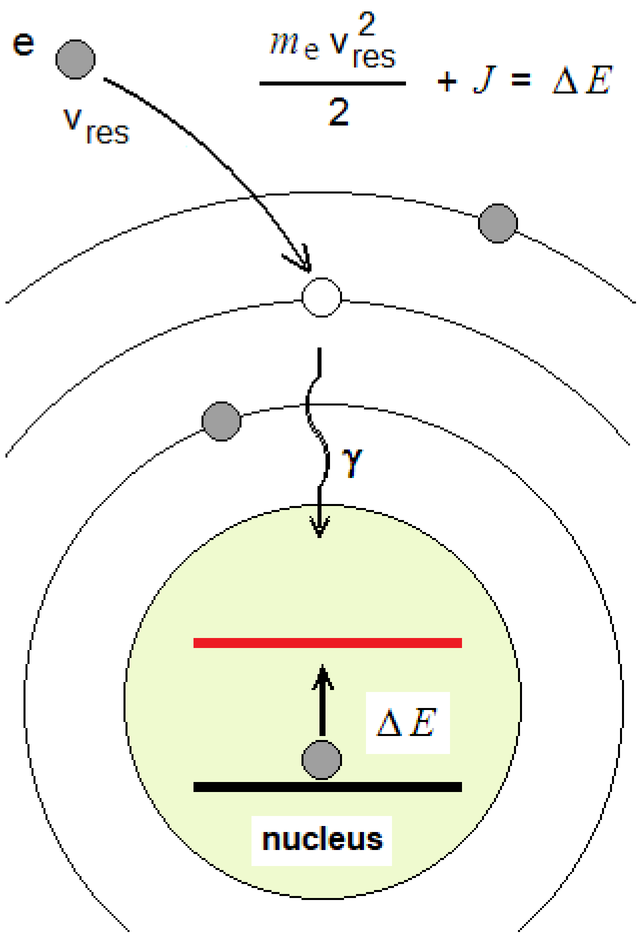
# De-excitation of nuclear isomers is most effective in plasma

In a plasma with isomeric nuclei at an electron temperature $\Theta_e \sim \Delta E$, the following are simultaneously present:

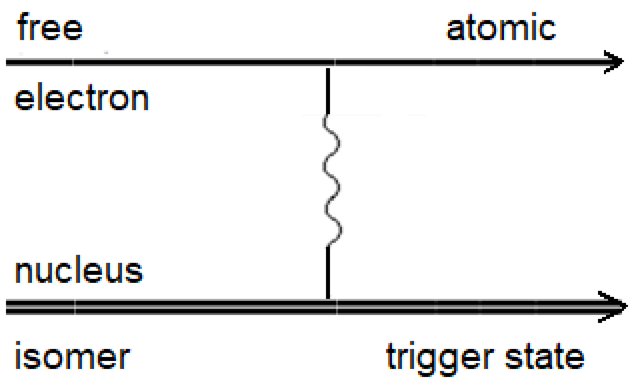
1. Intense X-ray radiation at the nuclear transition frequency.
2. Intense fluxes of electrons and ions.
3. High degree of ionization of atoms with isomeric nuclei.

Probability of de-excitation of nuclear isomers is proportional to the plasma lifetime.

## The effective mechanism for the excitation of a trigger nuclear level in plasma is the reverse internal electron conversion (RIEC) *(Gol'danskiy and Namiot, 1976 )*




*J* – electron binding energy in the atom.



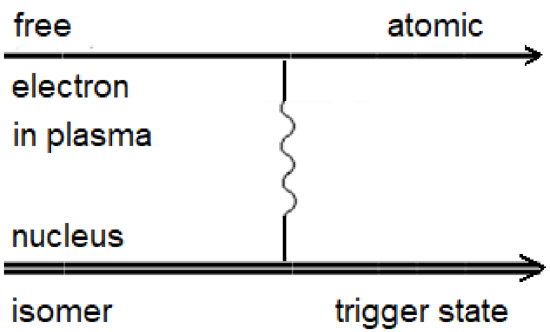

RIEC cross section

$$\sigma \sim \lambda_e^2 = (\hbar / m_e v_{res})^2$$

There is direct experimental confirmation of RIEC ( *Chiara, Carroll, Karamian, ..., 2018*).

## Excitation of a trigger level in plasma by the mechanism of reverse internal electron conversion (RIEC) (*Koltsov, 2018*)

$$P_{\text{RIEC}} \sim ƛ^2 \, \tau \, \text{v}_{\text{res}} \, n_{E_{\text{res}}} \, \Gamma$$




$m_e$, $\text{v}_{\text{res}}$ – mass and velocity of plasma e⁻, $m_e \text{v}_{\text{res}}^2/2 + J = \Delta E$

$J$ – the ionization potential of the atomic level capturing e⁻ ;

$n_{E,\,\text{res}}$ – energy density of e⁻ states, $E_{\text{res}} = m_e \text{v}_{\text{res}}^2/2$

$\Gamma$ – width of the conversion transition from the trigger level to the isomer ;

$\tau$ – plasma lifetime. RIEC cross section $\sigma \sim ƛ^2 = (\hbar / m_e \text{v}_{\text{res}})^2$

$$n_{E_{\text{res}}} = \frac{2}{\sqrt{\pi}} \frac{n\sqrt{E_{\text{res}}}}{\Theta_e^{3/2}} \, e^{-E_{\text{res}}/\Theta_e}$$

Maxwell-Boltzmann distribution for $n_E$,

$n$ – concentration e⁻, $\Theta_e$ – plasma temperature.

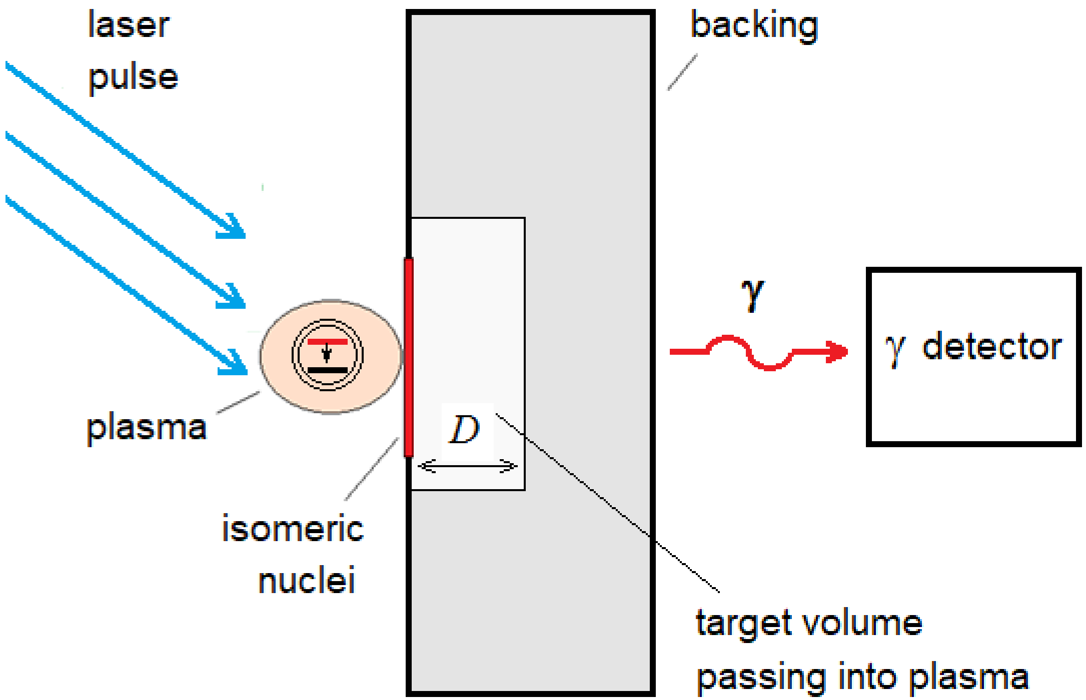
$$P_J \approx \frac{2\,g_i}{g_a} \left( \frac{m_e}{2\pi\hbar^2} \right)^{3/2} \frac{\Theta_e^{3/2}}{n} \, e^{-J/\Theta_e}$$

ionization probability of an atomic level $J$ according to the Saha formula.

$$P_{\text{excit.}} = P_{\text{RIEC}} \, P_J \sim \frac{1}{\pi^2} \frac{\Gamma \tau}{\hbar} \, e^{-\Delta E/\Theta_e}$$

Optimally: $\Theta_e \geq \Delta E$

# Experiments with laser plasma




Stimulated de-excitation of isomers can be detected

- by promt γ-quanta radiation ,
- with less sensitivity by nonequilibrium **α, γ,** $e^-$ radiation after de-excitation of the isomer.

Features of laser plasma:

1. The plasma lifetime is of the order of the laser pulse duration.
2. Depth D <1 μm, the number of isomer nuclei in plasma depends on the concentration of the isomer in the target material.

# Stimulation of $^{186m}$Re isomer de-excitation

(*V. V. Vatulin, N. V. Jidkov, A. A. Rimsky-Korsakov, Kolktsov, Kostylev, ..., 2017* )

Target camera of Iskra-5 laser facility

(*Institute of Experimental Physics, Sarov*).

Laser pulse :

- $\lambda$ = 1.3 μм,
- энергия ≈ 300 J,
- duration 0.3 ns,
- Intensity ~ $10^{15}$ W / cm$^2$.

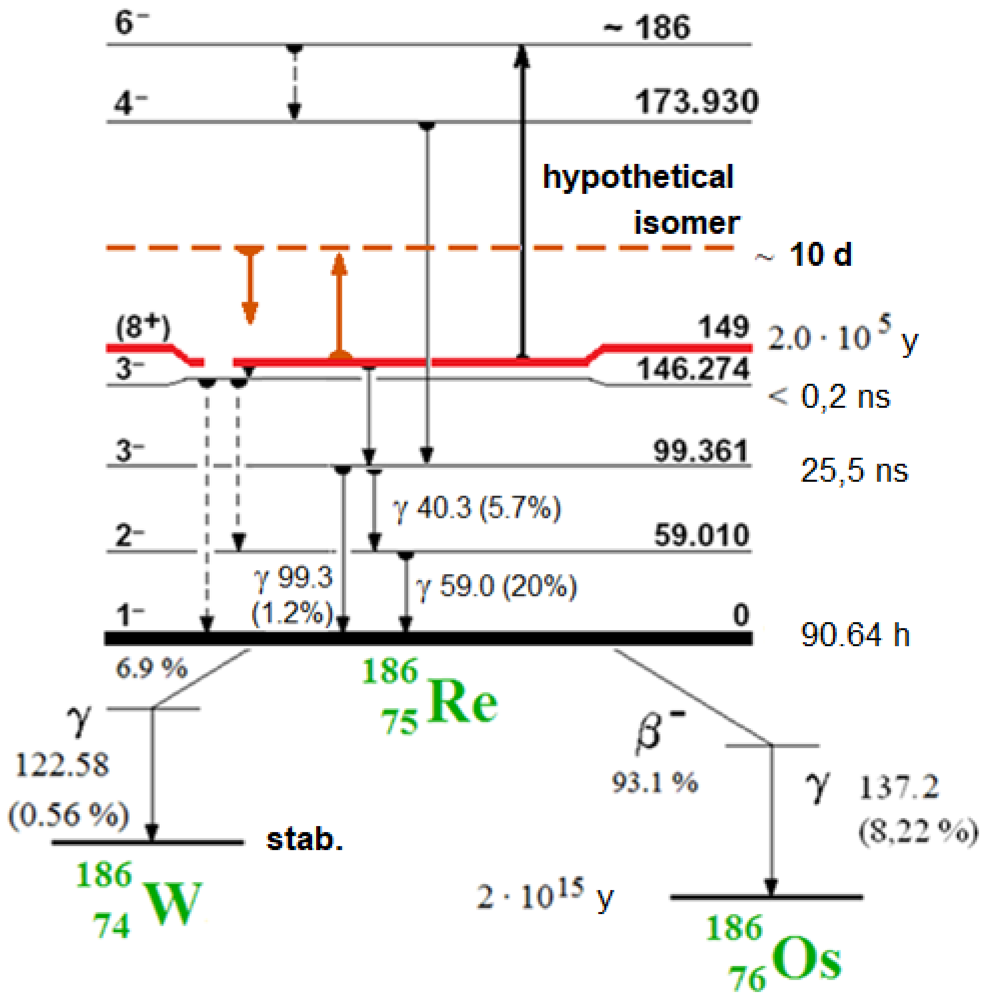
Plasma temperature $\Theta_e$ ~ 1 keV.

Targets:

- isomer 186mRe on W or Fe backings,
- isomer atomic concentration $^{186m}$Re ~ $10^{-3}$ %.

Isomer production - irradiation of a natural Re in reactor, flux ~ $1 \cdot 10^{20}$ neutron / cm$^2$.

# Isomer $^{186m}$Re is promising for energy source




Up arrows - possibility of exciting the trigger level.

Advanteges of the $^{186m}$Re isomer::

- Very long life time.
- Radiochemistry can isolate the pure 1$^{86m}$Re isomer.

Complicating circumstances:

- The trigger level parameters are unknown.
- The low accuracy ~ 1 keV of the isomer energy value prevents to resonantly affect the transition.

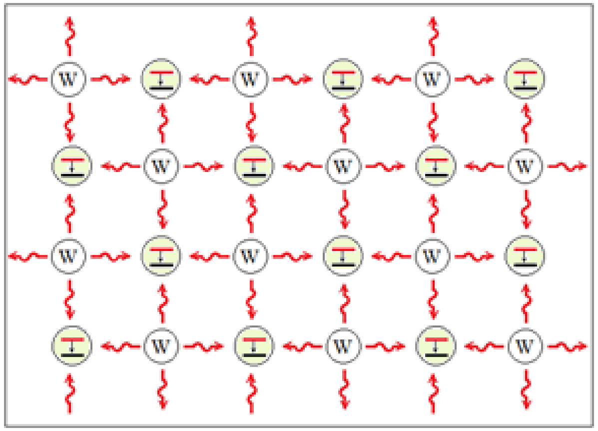
The known level diagram is incomplete. This was shown by measurements of the decay curve of $^{186}$Re, obtained in the reaction $^{186}$W (p , n) $^{186}$Re (*Koltsov, Rimsky-Korsakov, Karasev, 2018*).

## Ways to increase the probability of stimulated de-excitation of the $^{186m}$Re isomer in plasma

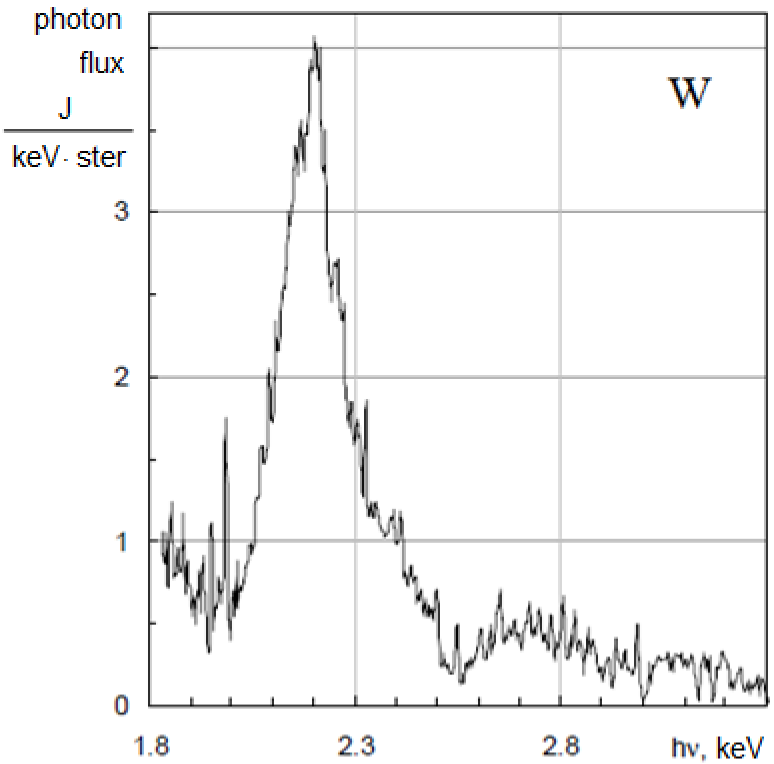
- Research of new mechanisms of trigger transitions stimulation. In particular, the study of the influence of resonant X-ray irradiation of frequency $\omega = \Delta E / \hbar$ on the probability of inverse internal conversion. For this, it is necessary to clarify the energy $\Delta E$ of the trigger transition.
- Increase of plasma lifetime.
- Increase of the concentration of isomeric nuclei in plasma.

# Amplification of RIEC with plasma irradiation by resonant photons of energy $\Delta E$ (1)

Resonant photon source – plasma self-radiation due to the introduction of atoms with a resonant X-ray characteristic line into the plasma together with atoms of $^{186m}$Re isomer.




Plasma from a mixture of isomer atoms and resonant emitter atoms (W).



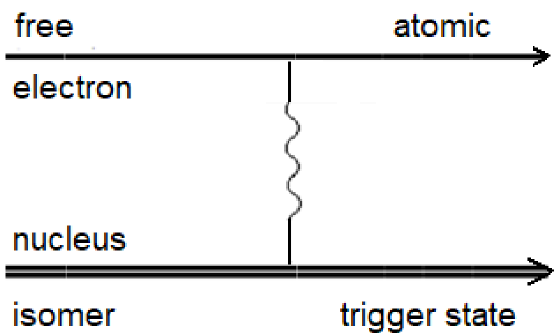

Example ( *Vatulin, ..., 2014* ):

spectrum of photons from plasma of the Iskra-5 facility, obtained by the action on the W-foil of a laser pulse of ~ 1 kJ, $\tau$ = 0.5 ns, intensity ~ $10^{16}$ W/cm$^2$.

Plasma temperature $\Theta_e$ ~ 1 keV.

≈ 1% of the laser pulse energy transfers into radiation of the W characteristic line.

# Amplification of RIEC with plasma irradiation by resonant photons of energy $\Delta E$ (2)




$$P_{\mathrm{RIEC}} \sim \lambda\!\!\!^{-2}\, v_{\mathrm{res}}\, \tau\, n_{\mathrm{res}}\ , \qquad n_{\mathrm{res}} = n \frac{\Gamma}{\Theta_e}$$

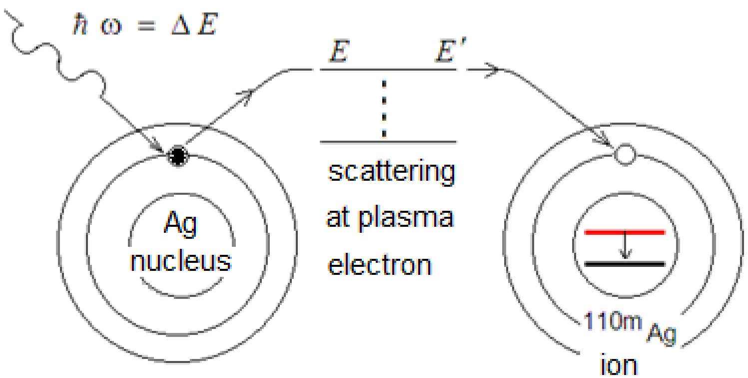
$n_{\mathrm{res}}$ – concentration of resonant electrons;

$\tau$ – plasma lifetime; $\Gamma$ – width of trigger transition;

$m$ , $v_{\mathrm{res}}$ – mass and velocity of plasma electrons; $m v_{\mathrm{res}}^2/2 + J = \Delta E$

$J$ – the ionization potential of the atomic level capturing $e^-$ .

1. Increasing the concentration of resonant electrons via the photoelectric effect (*Koltsov, 2019*)




$$P_{\mathrm{RIEC,res.}} \sim (1+\alpha)\, P_{\mathrm{RIEC}}$$

$\alpha$ depends on the plasma temperature $\Theta_e$ :

for $^{110m}$Ag isomer ( $\Delta E \approx 1.1$ keV )

$\alpha \sim 10^{-4}$ at $\Theta_e = 1$ keV, $\alpha \sim 10^{-2}$ at $\Theta_e = 10$ keV.

2. Stimulation of the virtual photon emission (*Koltsov, 2018*)

for $^{110m}$Ag isomer

$\alpha \sim 10^4$ at $\Theta_e \sim 10$ keV.

## Plasma of electric explosion of conductors – an alternative to laser plasma for trigger transitions energy ΔE up to ~ 1 keV (*Koltsov, 2018*)

Plasma expansion is slowed down by the magnetic field.

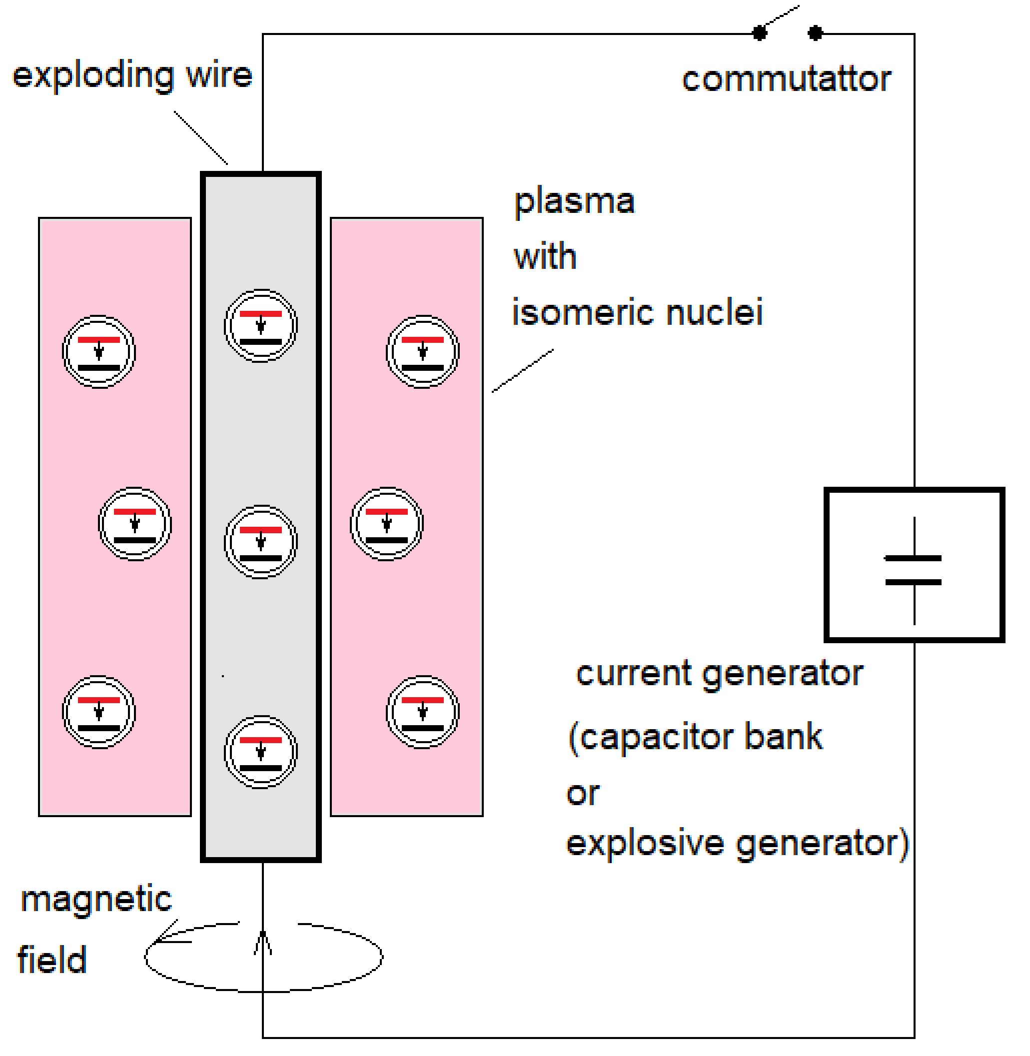
Attainable plasma parameters (*Angara-5M facility, Troitsk Institute for Fusion Research , TRINITI*) :

– temperature $\Theta_e$ ~ 1 keV;

– puls energy ~ 1 MJ;

– plasma lifetime $\tau$ = 10 – 100 ns.

"Exploding wire" is usually in the form of a liner - a set of wires for optimal plasma heating.




Scheme of electric explosion of conductors

# Advantages of stimulation of $^{186m}$Re isomers de-excitation in electro-discharge plasma

Higher efficiency of stimulation of de-excitation of isomers in comparison with laser plasma.

1. Due to increasing the plasma lifetime and a corresponding increase in the probability of stimulated de-excitation of isomers.
2. There is more matter in the electric discharge plasma than in the laser plasma. With one and the same isomeric material, the number of isomeric nuclei in the electric discharge plasma can be orders of magnitude larger than in the laser plasma.

# Angara-5 facility for producing the electro-discharge plasma

Troitsk Institute for Innovation and Fusion Research (TRINITI).

# “ Z ” facility

Sandia National Laboratories, USA.

# X-ray image unit

The view of a small-sized generator in an X-ray image unit produced by the Institute of High-Current Electronics (in Tomsk, 2008)

**Pulse generator:**

weight 70 kg, current up to 300 kA,

voltage 50 kV,

capacitors 250 nf,

current rise time 200 ns,

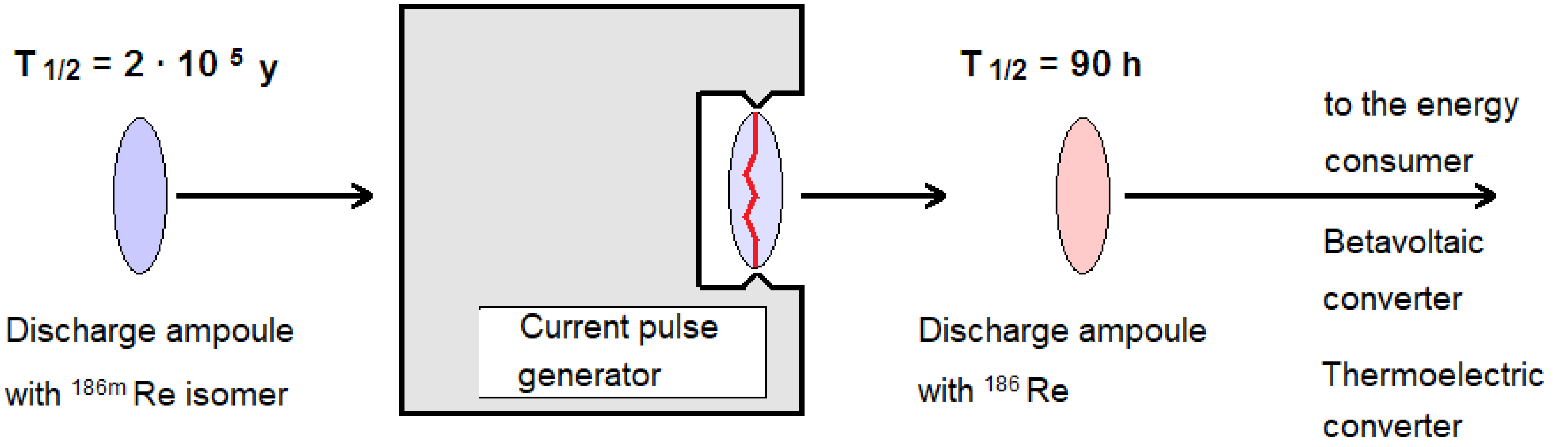
energy reserve of capacitor banks is 1 kJ.

X-pinch plasma

(electric explosion of crossed wires),

plasma temperature $\Theta_e$ ~ 1 keV.

# It is already possible to start applied work




The proposed scheme of the energy source.

A small-sized current pulse generator forms a plasma of the $^{186m}$Re isomer with a temperature $\Theta_e \sim 500$ eV in the discharge ampoule.

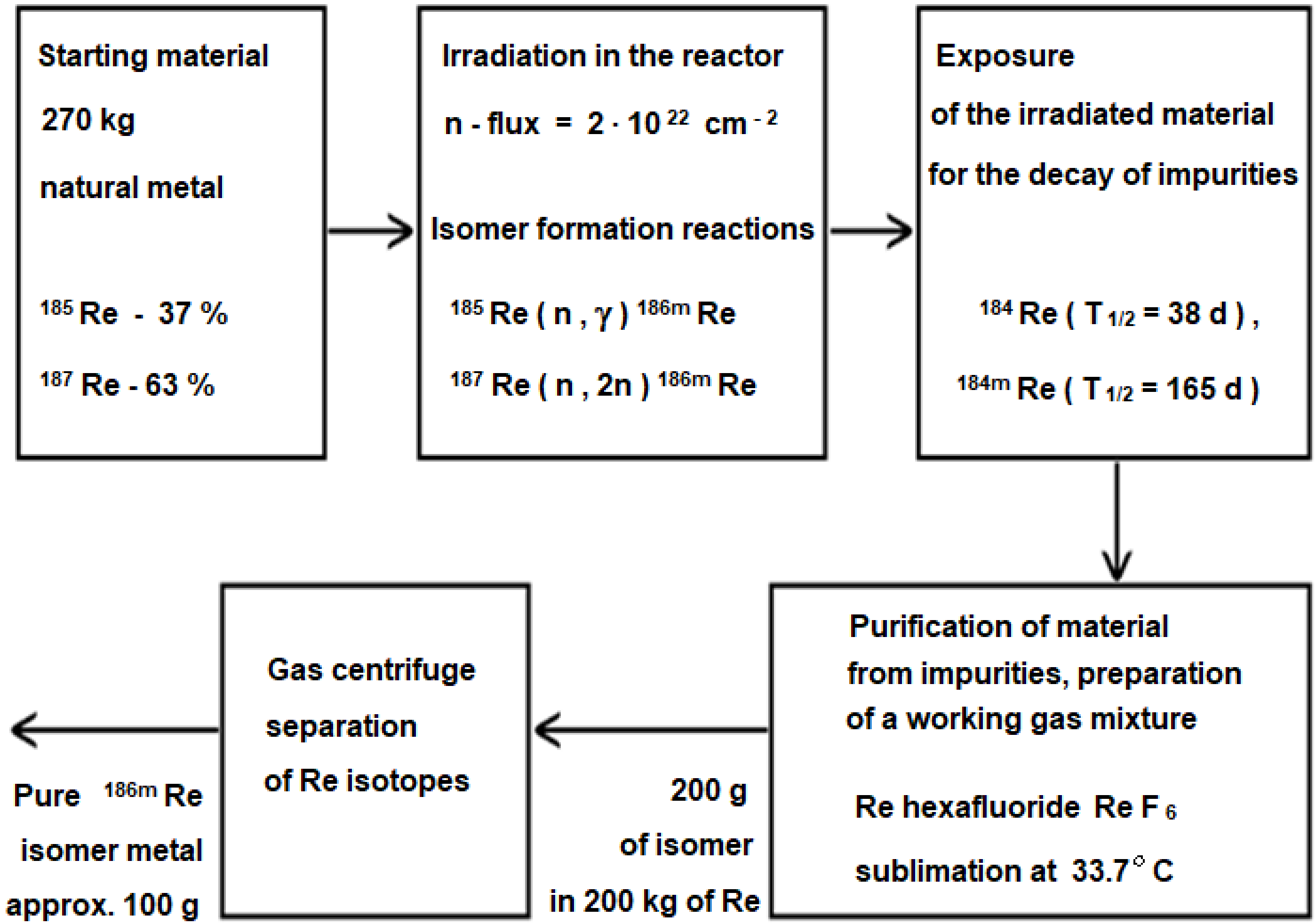
## Grounds for successful creation of $^{186m}$Re power source

- already obtained stimulated de-excitation of the $^{186m}$Re isomer in laser plasma,
- the outlined methods for enhancing the de-excitation of isomers in plasma,
- well-known technologies for the production of high current pulsed generators.

**Main problems to be solved:**

- determination of trigger transition parameters for the $^{186m}$Re isomer,
- experimental test of methods for increasing the probability of stimulating the de-excitation of the isomers in plasma,
- creation the technology for the production and isolation of large amounts of the $^{186m}$Re isomer.

# Scheme of the $^{186m}$Re isomer production



# Possible image of metal ingot from the pure isomer of rhenium

Isomeric metal is a new state of matter.

**Thanks !**