



РОСАТОМ



ГОСУДАРСТВЕННАЯ КОРПОРАЦИЯ ПО АТОМНОЙ ЭНЕРГИИ «РОСАТОМ»

Stimulation of ^{186m}Re isomeric nuclei de-excitation in high-current Z-pinch plasma at the “Angara-5” facility

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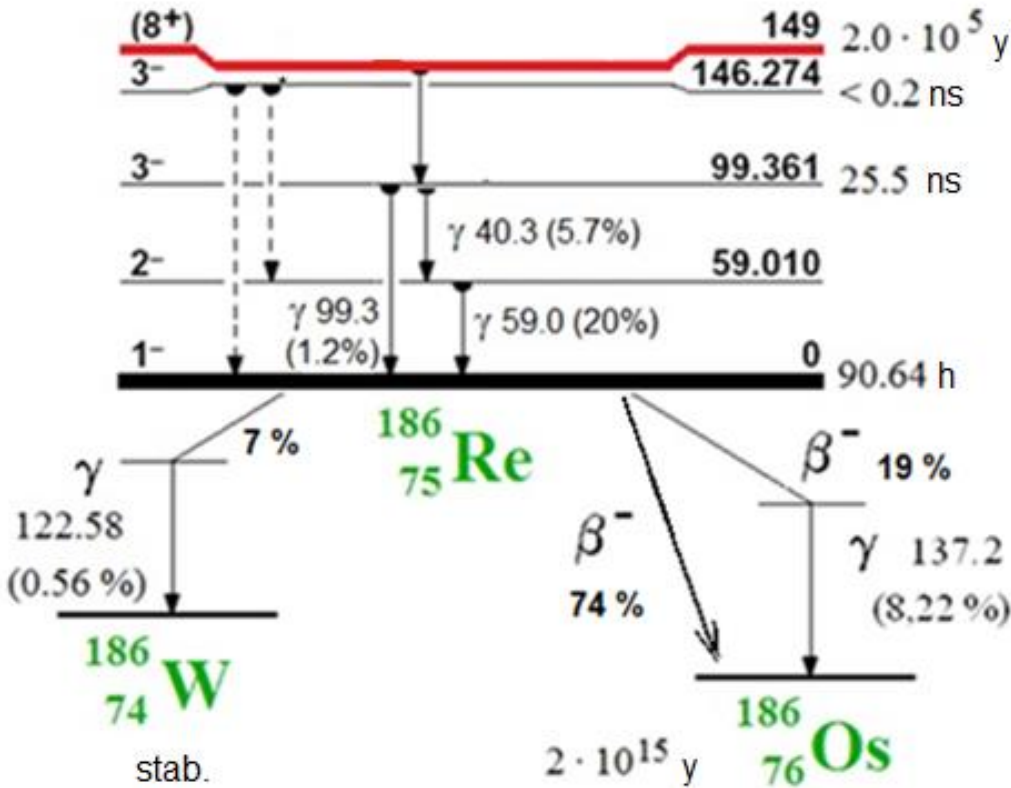
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^{186m}Re – Importance : the possibility of creating an energy accumulator based on the isomer



Battery operating principle:

- A working substance with ^{186m}Re isomeric nuclei is prepared, which can be stored indefinitely.
- To utilize the energy, deexcitation of the isomer in the plasma is stimulated.
- After this, energy of about 0.5 MeV is released per isomeric nucleus.

Discovery of the isomer : *Seegmiller 1972 [1]*.

The study of the ^{186m}Re isomer was proposed by V.N. Mikhailov (Sarov) in 2006, the work was organized by V.V. Vatulin (Sarov) and A.A. Rimsky-Korsakov (Radium Institute).



V.N. Mikhailov

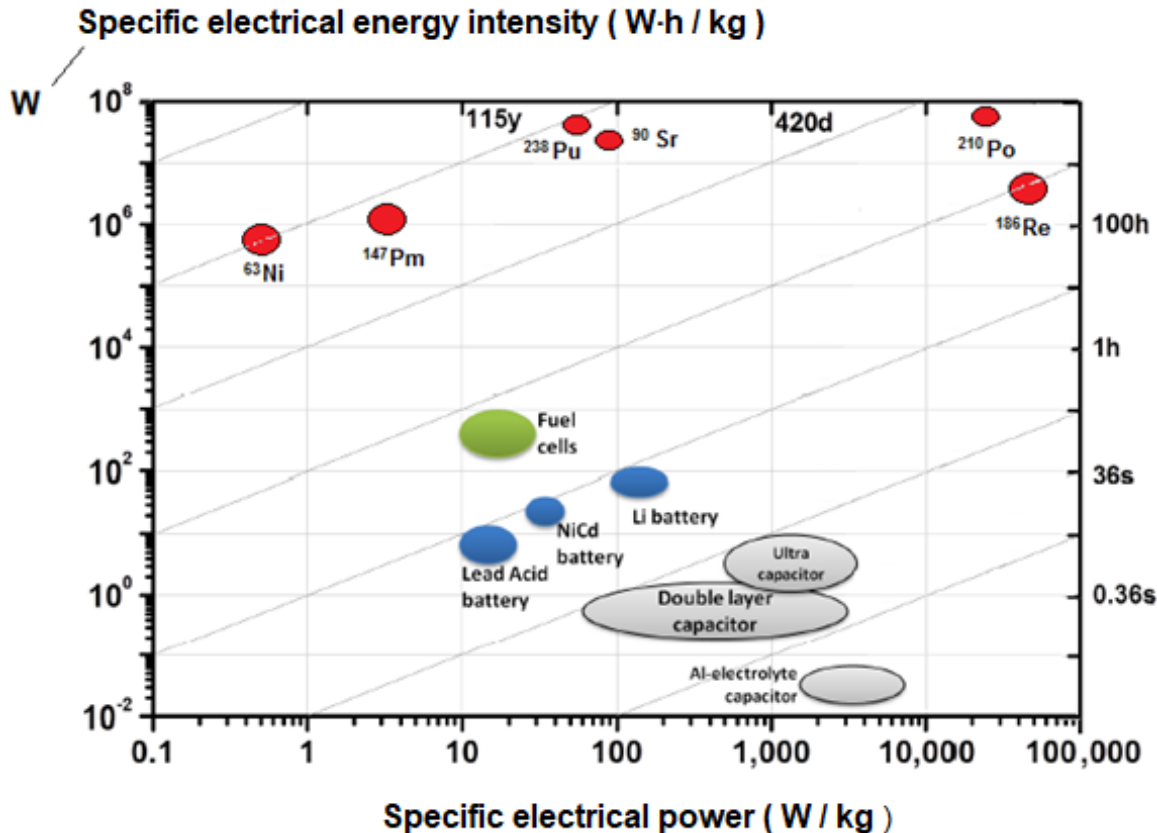


V.V. Vatulin



A.A. Rimsky-Korsakov

Comparison of radioisotope and non-nuclear electric batteries



- Battery based on ^{186m}Re isomer :
 - electrical power 100 W/g,
 - energy intensity 108 J/g,
 will outperform all known power sources.
- The cost of the ^{186m}Re isomeric material is comparable to the cost of the same mass of ^{238}Pu used today in energy sources.
- The ^{186m}Re battery, taking into account its relative environmental safety, will have a competitive advantage in the market over its analogues.

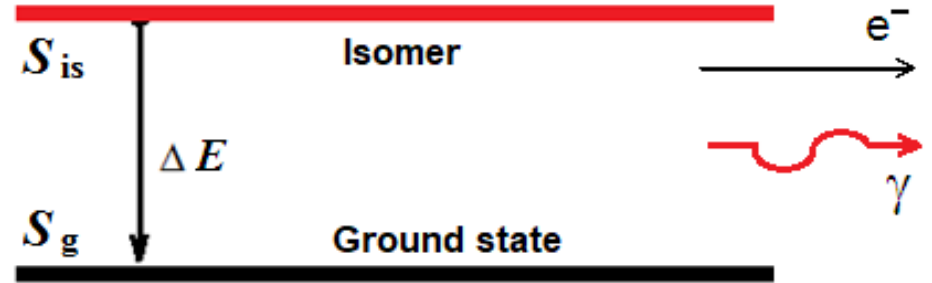
- The efficiency of converting the energy of α , β -particles into electrical energy is assumed to be 10%, radionuclides are assumed to be in the form of metal.
- Parameters of non-nuclear batteries from work : Zhou, Zhang, Wang ..., 2021 [2].

Outline

1. Possible mechanisms of deexcitation of nuclear isomers (SDENI).
2. Studies with isomeric ^{186m}Re nuclei in laser plasma.
3. Advantages of electric-discharge plasma of the Angara-5 facility for deexcitation of ^{186m}Re isomers.
4. Experimental technique for producing electric-discharge plasma with isomeric ^{186m}Re nuclei.
5. Deexcitation of the ^{186m}Re isomer in the plasma of the Angara-5 facility.

Spontaneous nuclear 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 of S_{is} and S_g .



1. Probability of emission

of a γ -quantum with wavelength λ

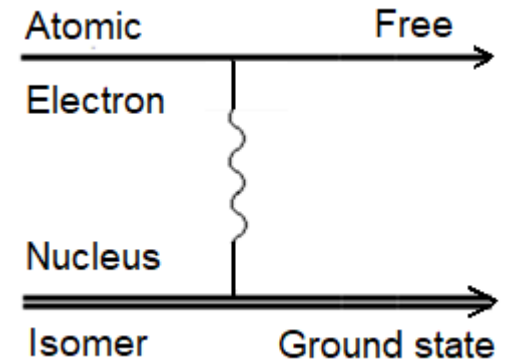
$$P_{\text{rad}} \sim \alpha \frac{c}{\lambda} \left(\frac{R_{\text{nucl}}}{\lambda} \right)^{2L}$$

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

2. Probability of conversion

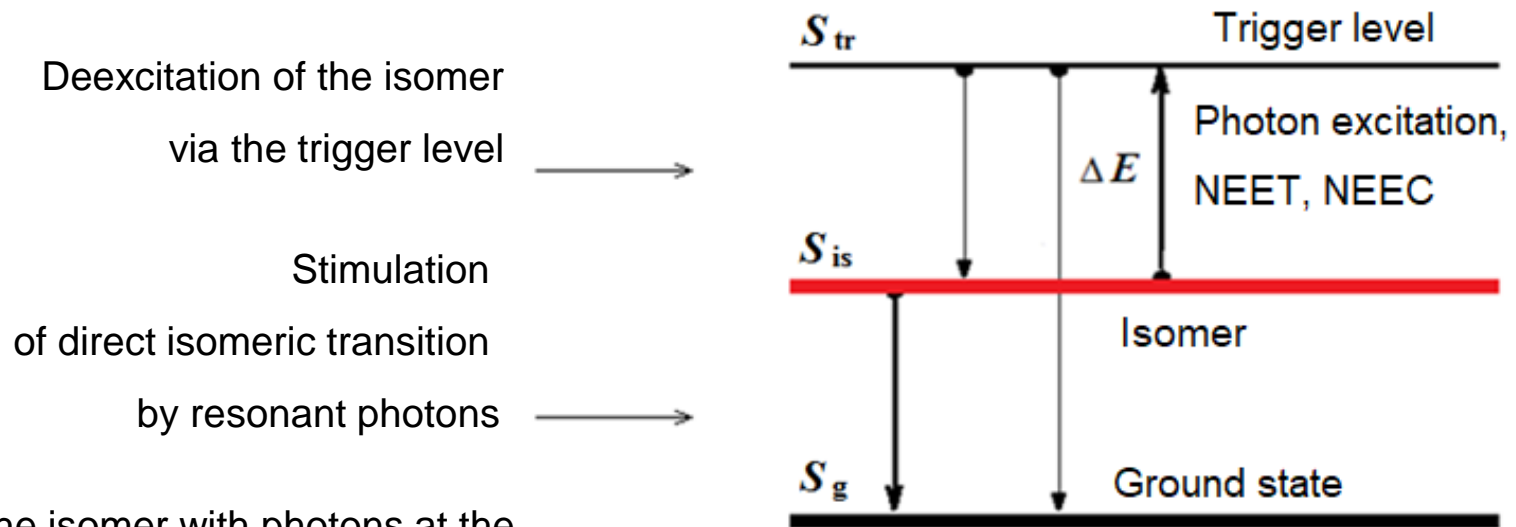
$$P_{\text{conv}} = \beta_{\text{conv}} P_{\text{rad}}$$

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



For low-energy EM transitions, electronic conversion is more probably and vice versa – the excitation of the transition is more effective via the excitation of the electron shell of the atom.

Methods for stimulating deexcitation of nuclear isomers (SDENI)



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);
- trigger level excitation.

2. Trigger level excitation:

- resonant absorption of photons;
- inelastic electron scattering;
- transfer of energy from the atomic shell to the nucleus (NEET, NEEC).

- For a trigger transition, the multipolarity is less than for an isomeric transition::

$$S_{is} > S_{tr} > S_g$$

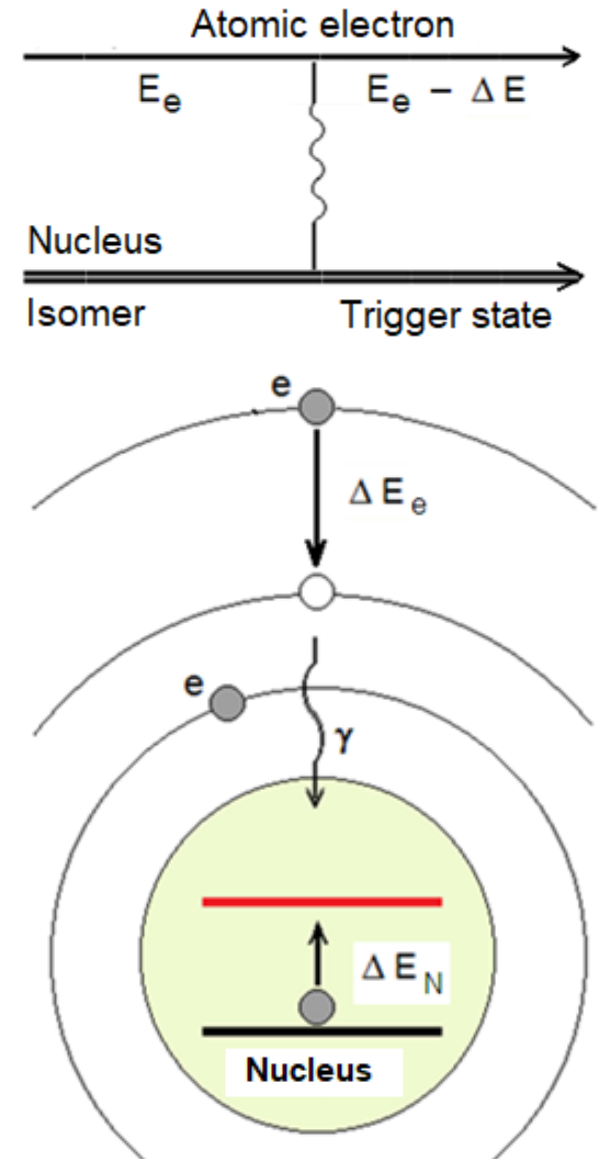
- In practically important cases $\Delta E > 1$ keV.
- Not all possible trigger levels are known at this time.

Methods to excite close trigger levels

1. Excitation of the isomeric nucleus by photons $\hbar\omega = \Delta E_N$ (for radiation in plasma: *Letokhov, 1973 [4]*).
 2. Inelastic scattering of electrons on an isomeric nucleus (for the ^{235m}U isomer: *Grechukhin and Soldatov, 1976 [5]*).
 3. Nuclear excitation by electron transition (NEET) is the transfer of excitation to the isomeric nucleus via an electronic transition between atomic levels (*Morita, 1973 [6]*).
- Resonance between atomic and nuclear transition is needed $|\Delta E_N - \Delta E_e| < \Gamma_e$ – the atomic transition widths.

Achieving resonance for an excited atom with an isomeric nucleus:

- Photon irradiation $\hbar\omega = \Delta E_e - \Delta E_N$ (*Zon, Karpeshin 1990 [7]*).
- Deformation of the electron shell of isomeric nuclei in atomic collisions (*Koltsov 2023 [8]*).

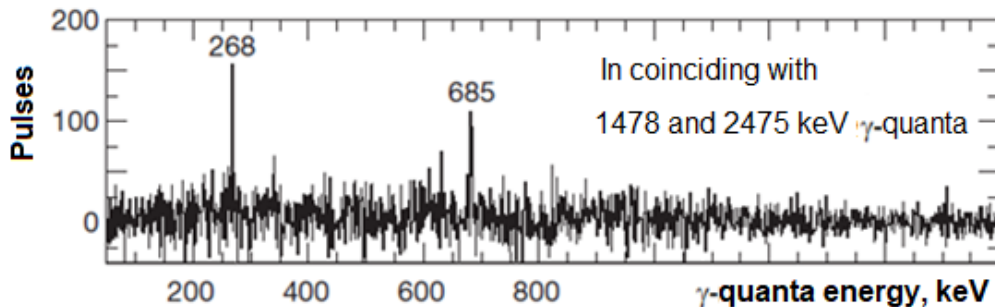
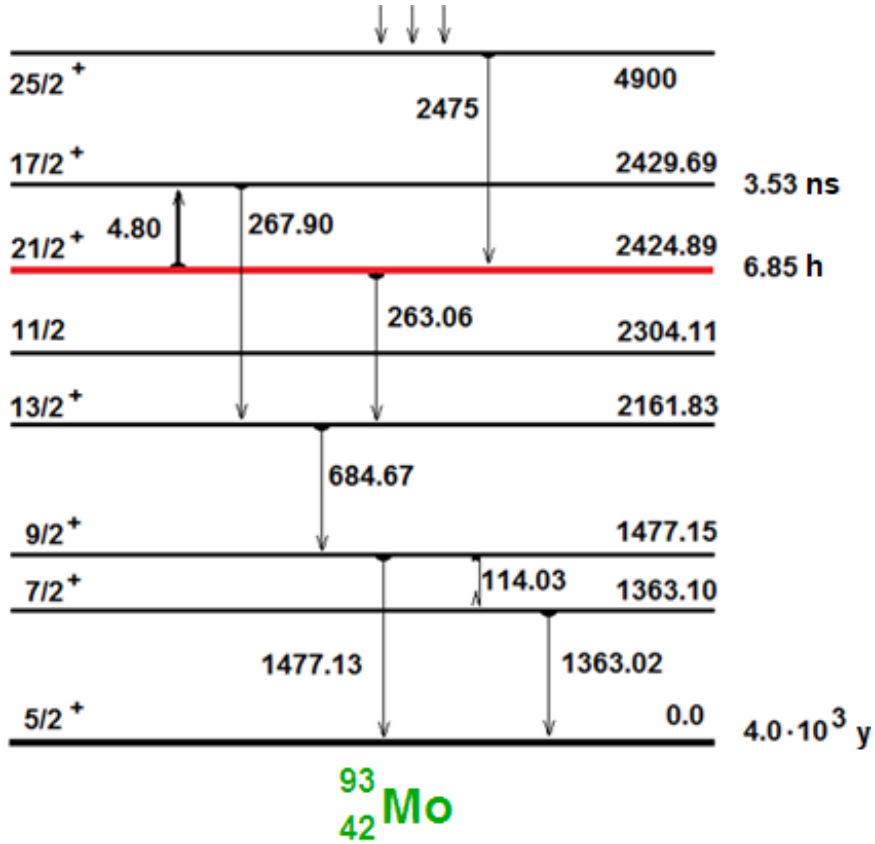


NEET scheme: needed $\Delta E_e \approx \Delta E_N$

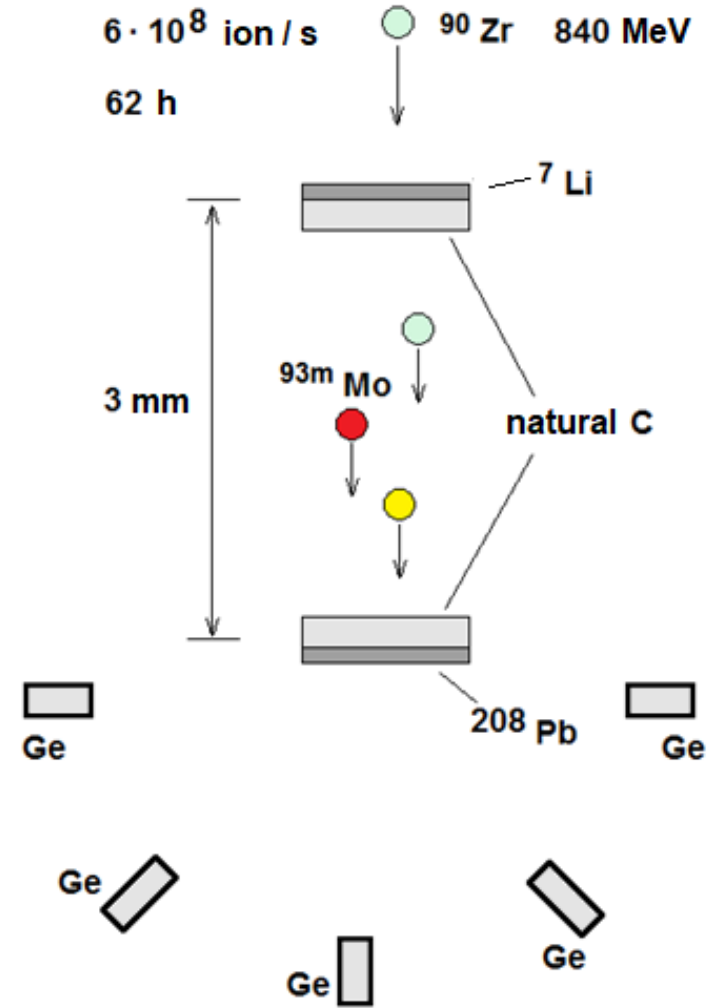
Excitation of the trigger level in the ^{93m}Mo

(Chiara, Carroll, Karamian, ..., - 2018 [9])

After evaporation of nucleons from $^{90}\text{Zr} + ^7\text{Li}$



ATLAS facility (Argonne National Laboratory)



92 γ -detectors in coincidence

The probability of a trigger transition is much greater than via the NEEC mechanism (Wu, Keitel, Palffy ... - 2019 [10]).

Promising isomers $T_{1/2} > 3$ days

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

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

- (pink) – de-excitation was stated when irradiating isomers with photons, there is no confirmation.
- (blue) – deexcitation of isomers in plasma.

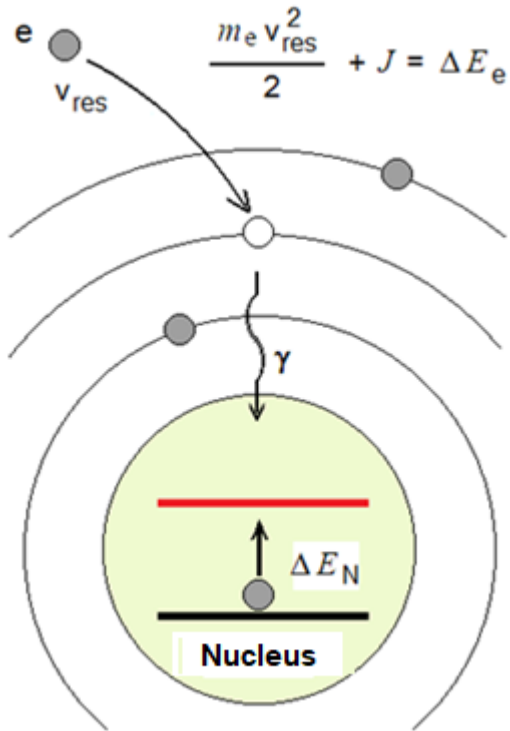
Excitation of atomic nuclei in plasma

In a plasma with isomeric nuclei at an electron temperature on the order of the nuclear transition energy $\Theta_e \sim \Delta E_N$, the following are simultaneously present:

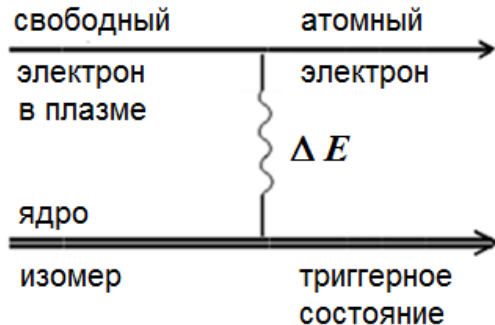
- Intense *X*-ray radiation at the frequency of the nuclear transition.
- Intense flows of electrons and ions.
- High degree of ionization of atoms.
- Deformation of electron shells of nuclei in atomic collisions.

Probability of nucleus excitation in plasma
is proportional to the plasma lifetime.

Nuclear excitation by plasma electron capture (NEEC) (Goldansky, Namiot, 1976 [12])



J – энергия связи электрона.



When $\Delta E_e = \Delta E_N$ the nucleus is excited.

NEEC cross section $\sigma_{рез} \sim \tilde{\lambda}_e^2 = \left(\hbar / m_e v_{рез} \right)^2$

NEEC probability $P_{ОВЭК} \sim \tilde{\lambda}^2 \tau v_{рез.} n_{E_{рез.}} \Gamma$

m_e, v_{res} – mass and velocity of e^- plasma, $m_e v_{res}^2 / 2 + J = \Delta E$

$n_{E, res.}$ – energy density of e^- – states, $E_{res.} = m_e v_{res.}^2 / 2$

Γ – the width of the conversion transition from the trigger level to the isomer;

τ – plasma lifetime.

Estimation of the excitation probability in plasma (Koltsov, 2021 [13])

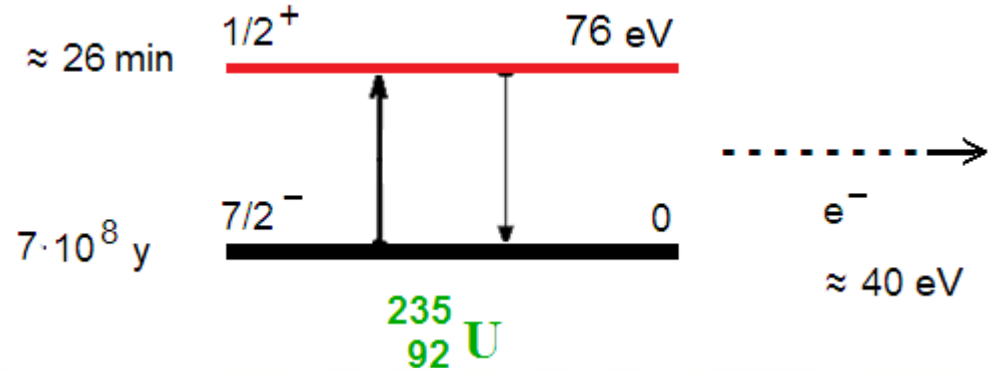
$$P_{excit} \sim \frac{1}{\pi^2} \frac{\Gamma \tau}{\hbar} e^{-\Delta E_N / \Theta_e} \quad \text{Optimally :} \quad \Theta_e \geq \Delta E_N$$

Θ_e – plasma electron temperature.

Excitation of the ^{235m}U isomer in plasma (TRINITI)

(Arutyunyan, Bolshov, Koltsov, Malyuta, Rimsky-Korsakov, Smirnov, Tkalya ... 1989 [14])

Target – oxide ^{235}U
(ground state)

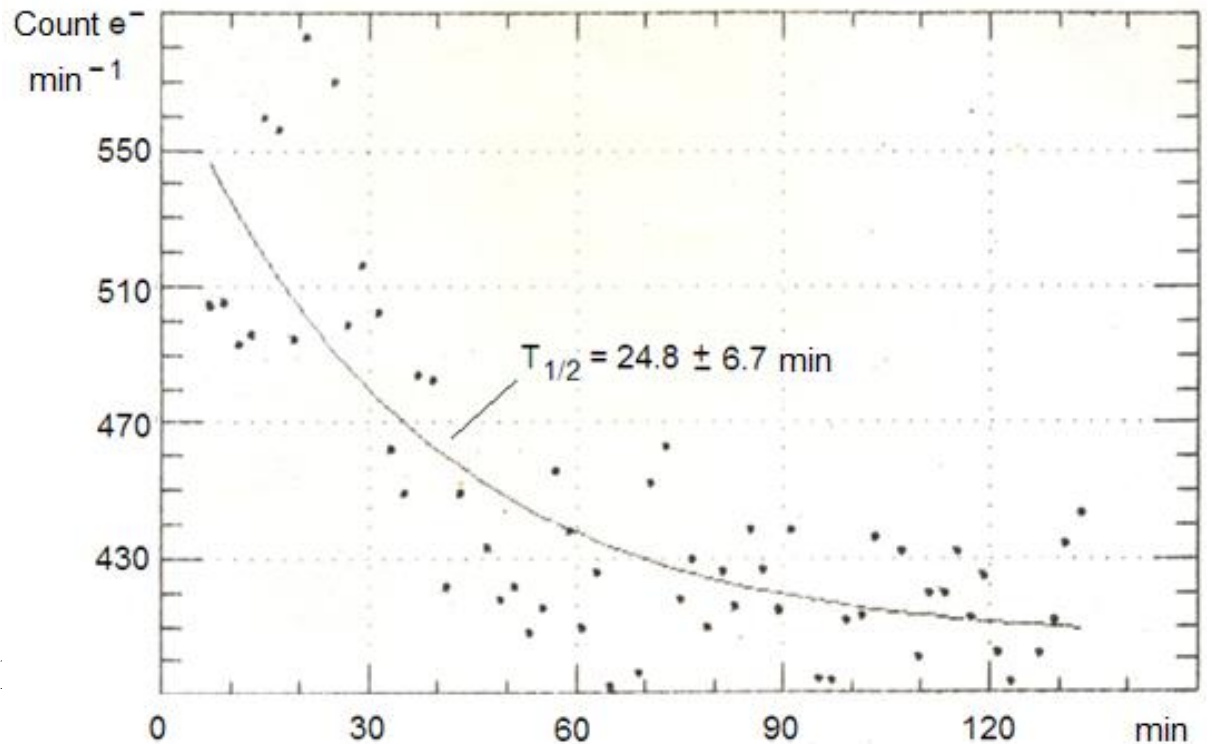


Plasma from an electron beam:

- pulse duration 30 ns,
- current 150 kA,
- e^- energy 500 keV,
- plasma spot diameter 2 mm,
- plasma volume $\sim 10^{-4} \text{ cm}^3$.

Plasma temperature $\Theta_e \sim 20 \text{ eV}$.

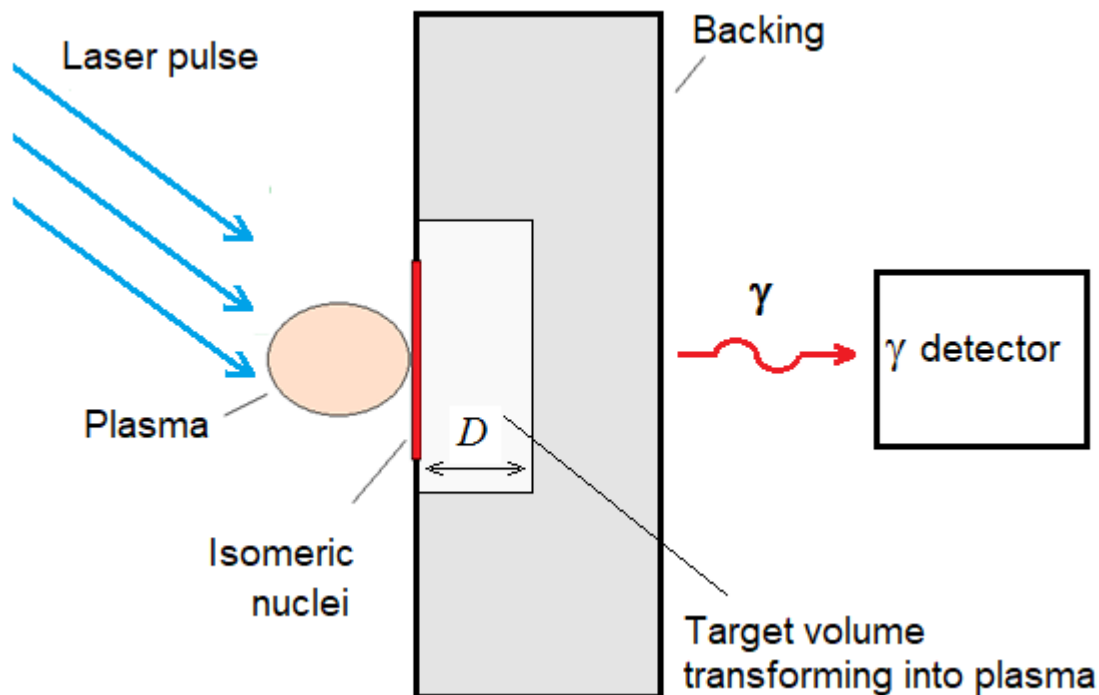
$$P_{\text{возб.}} \sim \frac{1}{\pi^2} \frac{\Gamma \tau}{\hbar} e^{-\Delta E / \Theta_e} \sim \dots$$



The estimate coincides with the experiment.

e^- conversion intensity after electron shot.

Scheme of SDENI experiments with laser plasma



Stimulated deexcitation of isomers can be detected :

- by instantaneous g-quanta;
- with less sensitivity by nonequilibrium radiation α , γ , e^- from a sample of plasma matter after deexcitation of the isomer.

Specifics of laser plasma:

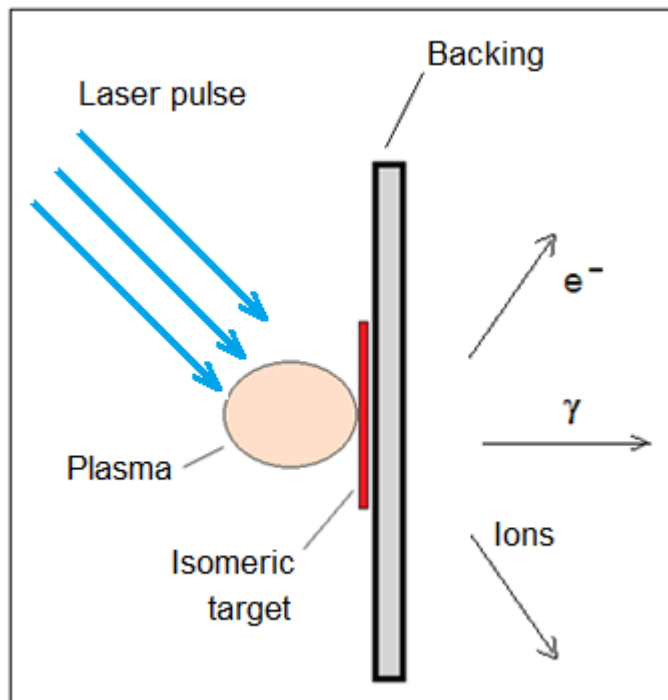
- The plasma lifetime is on the order of the laser pulse duration.
- Depth $D < 1 \mu\text{m}$.
- The number of isomer nuclei in the plasma depends on the concentration of the isomer in the target material.

Plasma of high-power ultrashort laser pulses

Laser pulse:

- duration < 1 ps
- energy ~ 100 J
- intensity $> 10^{18}$ W / cm²
- target volume $< 10^{-7}$ cm³.

Plasma temperature up to 10 keV.



Features of the plasma:

- Nonequilibrium e⁻ of energy up to ~ 10 MeV.
- X-ray up to ~ 100 MeV.
- High energy ions ($E_p > 10$ MeV).

The following was observed in the plasma:

- photonuclear reactions (γ, n),
- ²³⁵U photo fission,
- nuclear reactions (p, n),

– excitation of nuclear transitions :

- ¹⁸¹Ta ($E1$ transition $\Delta E = 6.2$ keV)
(Andreev, Gordienko ..., 2000 [15]);
- ¹⁰³Rh ($E3$ transition $\Delta E = 39.8$ keV)
(Afonin, ..., 2012 [16]).

It is difficult to hope on creating an energy source based on photonuclear and (p, n) reactions due to the small cross sections and short plasma lifetime.

Stimulation of deexcitation of the ^{186m}Re isomers

(Vatulin, Zhidkov, Rimsky-Korsakov, Koltsov, Tachaev, ..., 2017 [17])

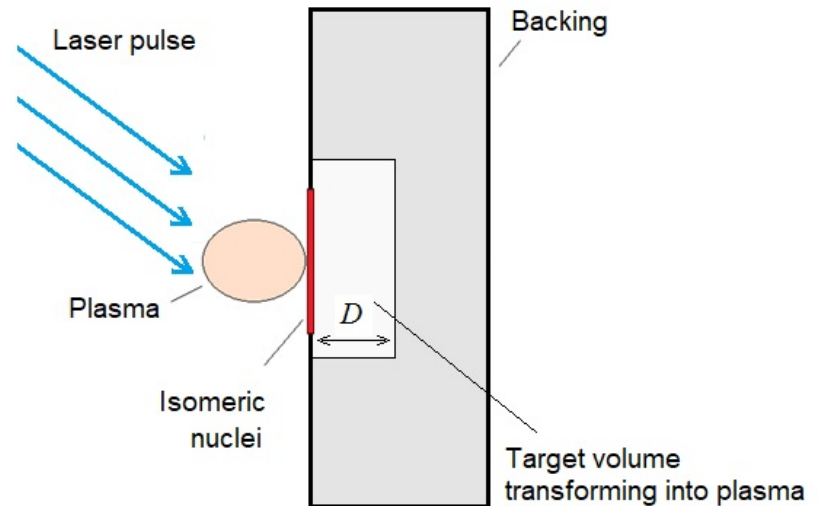


Target camera of the Iskra-5 laser system
(Institute for Experimental Physics – Sarov).

Laser pulse:

- $\lambda = 1.3 \mu\text{m}$,
- Energy $\approx 300 \text{ J}$,
- Duration 0.3 ns ,
- Intensity $\sim 10^{15} \text{ W / cm}^2$.

Plasma temperature $\Theta_e \sim 1 \text{ keV}$.

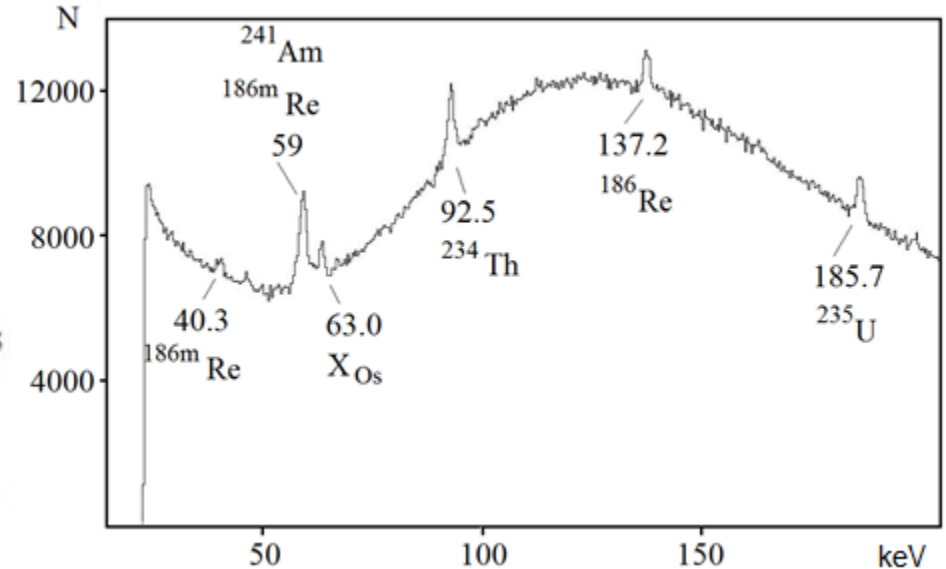
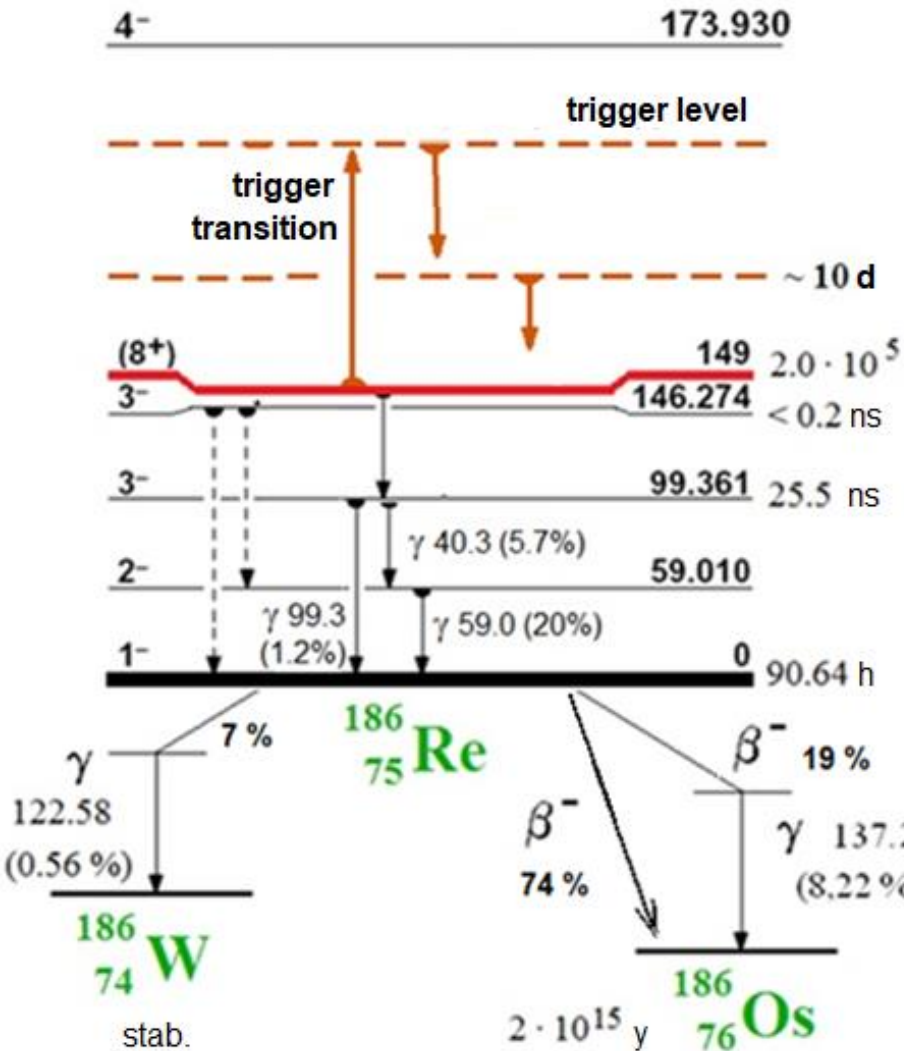


Experimental scheme. The amount of isomer in plasma depends on its concentration in the target.

Targets: ^{186m}Re isomer on W or Fe backings.

- In an isomeric material, the atomic concentration of the ^{186m}Re isomer is $\sim 10^{-3} \%$.
- Irradiation in a reactor of natural Re , fluence $\approx 6 \cdot 10^{19} \text{ neutron / cm}^2$.

Method for detecting stimulation of deexcitation of the ^{186m}Re isomer in plasma

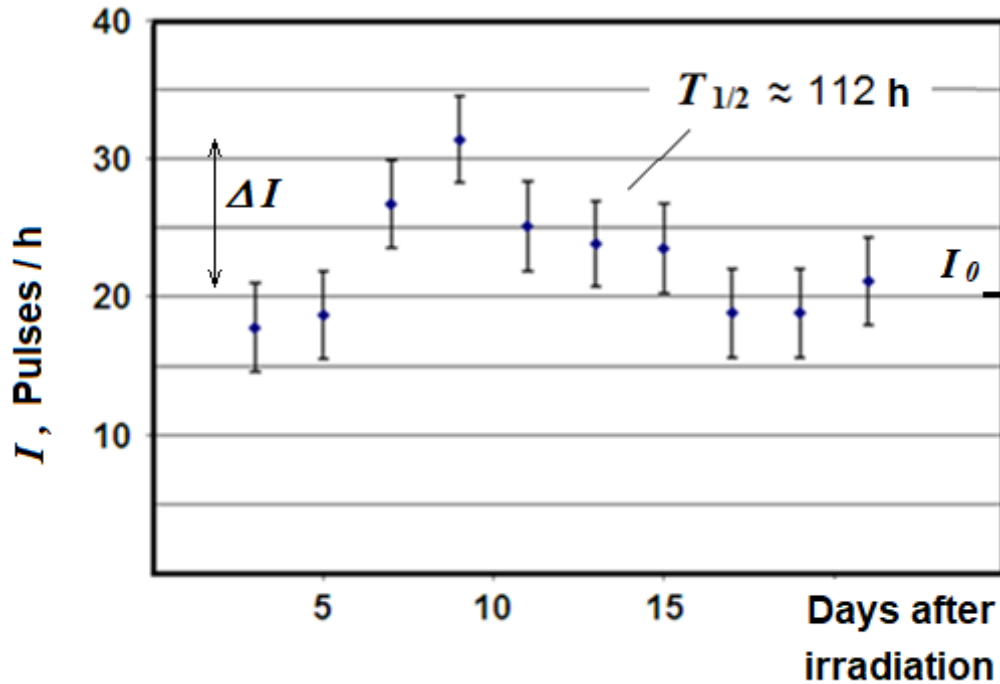


γ -spectrum of a plasma matter sample:

- N – number of readings over 8 days of measurements.
- Activity of ^{186m}Re in the sample is ≈ 0.1 Bq.
- γ -detector – HPGe 150 cm^3 with a well.
- The additional intensity of 137 keV γ -quanta decreases with the half-life of the levels populated in the decay of the trigger level.

- With pulse stimulation of isomer deexcitation, the population of the trigger level increases stepwise.

Time variations of the intensity of the 137 keV γ -line in the spectrum of irradiated ^{186m}Re



The result of one of the experiments on a laser target with a W-backing.

- Exposure of each point is 2 days.
- The activity of the plasma sample ≈ 0.1 Bq.
- The decrease in intensity corresponds to $T_{1/2} = 112 \pm 10$ hours.
- The effect was not observed on targets with a steel backings.

If the effect is due to the stimulated deexcitation of nuclei ^{186m}Re , then:

- The presence of a maximum shows that in the ^{186}Re nucleus there is an unknown isomer with $T_{1/2}$ for several days, which is populated in stimulated discharge of the known ^{186m}Re isomer.
- Probability of deexcitation of nuclei ^{186m}Re :
$$P_{\text{сДЯИ}} = \frac{N_{\text{сДЯИ}}}{N_{\text{is}}} = \frac{\Delta I}{I_0} \frac{T_{1/2}}{T_{\text{is}}} \approx 1 \cdot 10^{-7}$$
- The trigger level for ^{186m}Re is still unknown \Rightarrow it is unclear how the effect was obtained.

Search for the ^{186m}Re isomer deexcitation by bremsstrahlung radiation up to 40 keV. No effect.

- We tried to clarify the mechanism of deexcitation of the ^{186m}Re isomer by irradiating the isomer with photons and electrons without the formation of plasma.
- At the same time, the integrated fluxes of electrons and photons far exceeded the corresponding fluxes in the plasma of the Iskra-5 facility.

$$V_e = 46 \text{ kB,}$$

$$I_e = 10 \text{ mA.}$$

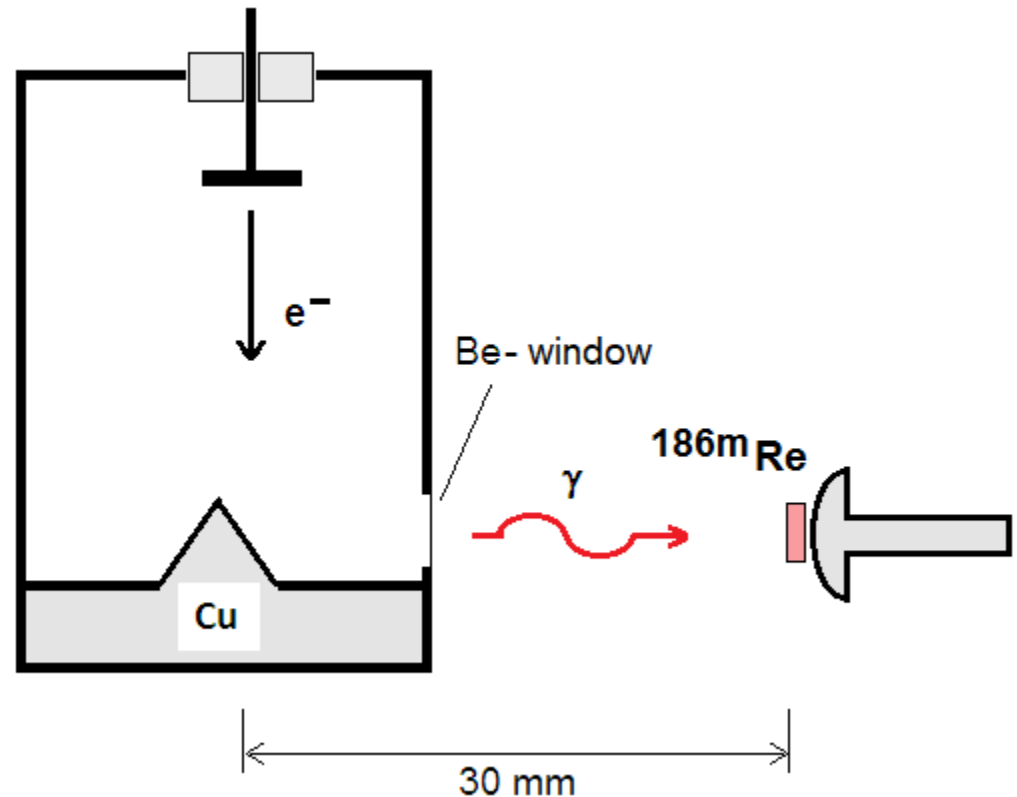
Irradiation exposure 400 minutes.

- Re metal layer with isomer:
thickness 5 microns, \varnothing 4 mm.

- Photons:

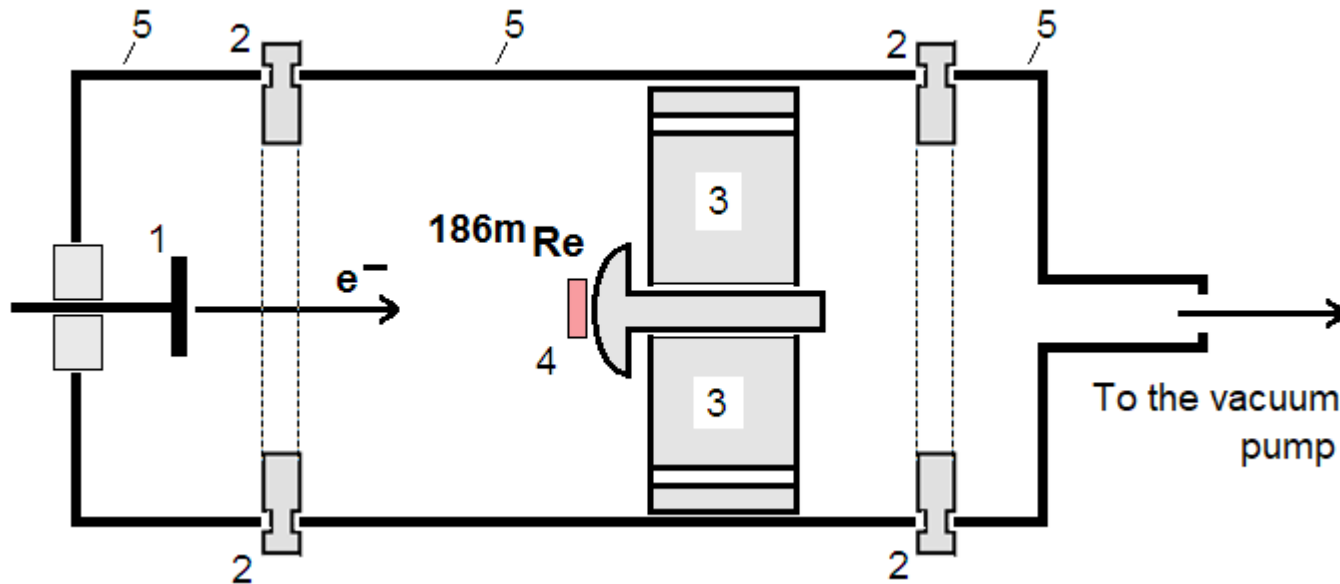
$$E = 20 \text{ keV,}$$

$$\text{flux} = 10^{14} \text{ photons / (keV} \cdot \text{cm}^2\text{).}$$



X-ray tube BS V-9.

Search for the ^{186m}Re isomer deexcitation by 10 keV electrons. No effect.



- $E_e = 10 \text{ keV}$, $I_e = 3 \text{ mA}$, Beam section 10 mm.
- Re metal layer 1 μm thick, $\varnothing 4 \text{ mm}$.
- Irradiation exposure is 150 minutes.
- Bremsstrahlung photons $E = 3 \text{ keV}$:
flux = $4 \cdot 10^{17}$ photons / (keV \cdot cm²).

- 1 – electron emitter (cathode);
- 2 – electrically insulating flanges;
- 3 – massive copper target holder;
- 4 – ^{186m}Re on a stainless steel backing in the form of a bolt;
- 5 – electron gun body.

No effect. **Conclusion: deexcitation of the ^{186m}Re isomer occurs only in plasma.**

Possible ways to increase the probability of stimulating de-excitation of nuclear isomers (SRENI) in plasma

To create an energy accumulator, it is necessary to increase the efficiency of SDENI in plasma.

This requires new experimental techniques:

- Increased concentration of isomeric nuclei in plasma.
- Clarification of the scheme of trigger transitions in the studied isomers.
- Study of stimulation mechanisms, in particular, study of the influence of X-ray plasma irradiation, resonant with trigger transitions, on the probability of SDENI.
- Search for new nuclei convenient for research with a more accurately known trigger transition energy.
- Increased plasma lifetime.

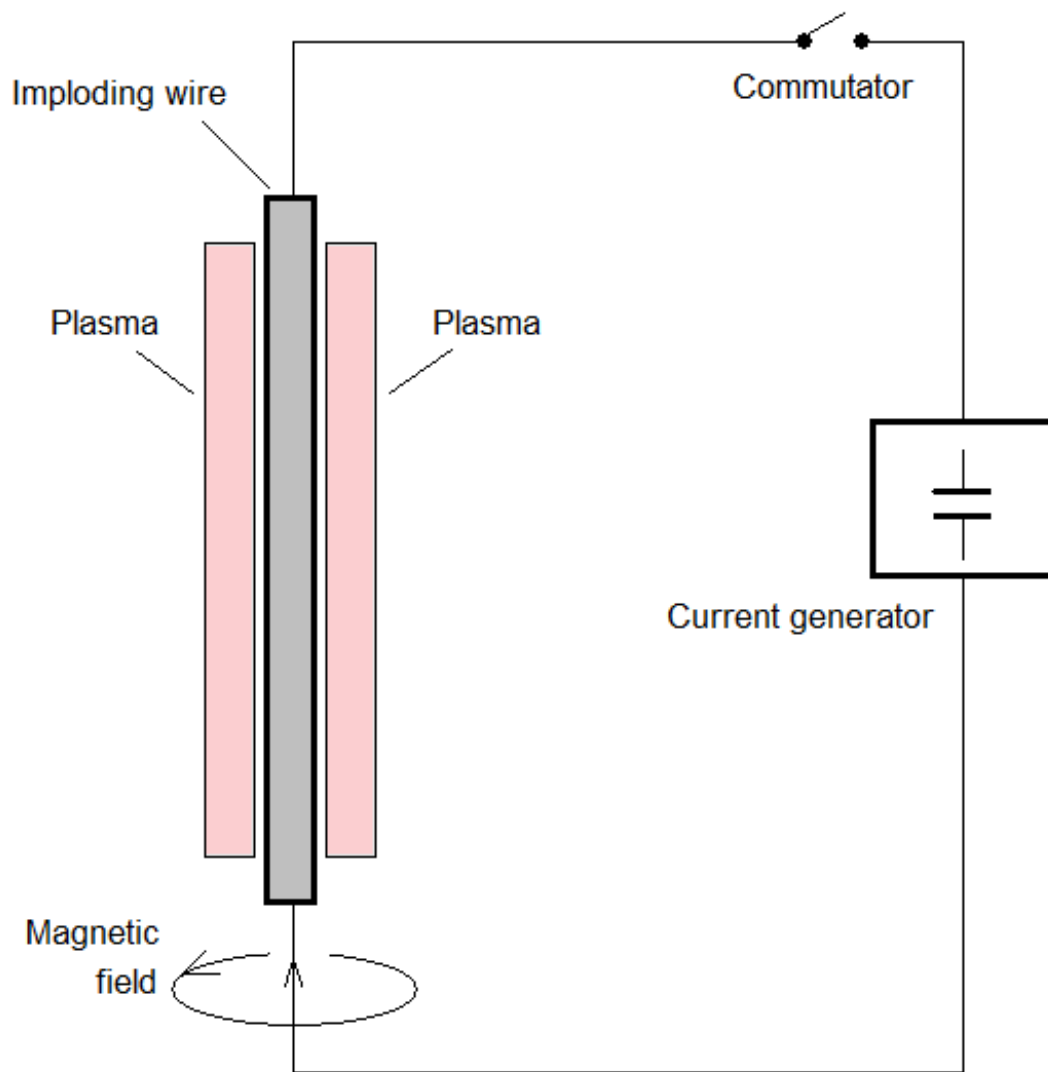
**Plasma of electrical implosion of conductors (EICP) is an alternative to laser plasma
for isomeric transitions of energy ΔE up to ~ 1 keV (Koltsov, 2021 [13])**

Achievable plasma parameters
of the Angara-5m facility,
“TRINITI” (Troitsk):

- Temperature Θ_e up to ~ 1 keV;
- Energy per pulse up to ~ 1 MJ;
- Lifetime $\tau = 10 - 100$ ns.

The expansion of the plasma is slowed
down due to the magnetic confinement
of the plasma.

“Imploding wire” usually in the form of
a liner - a set of wires for optimal
plasma heating.



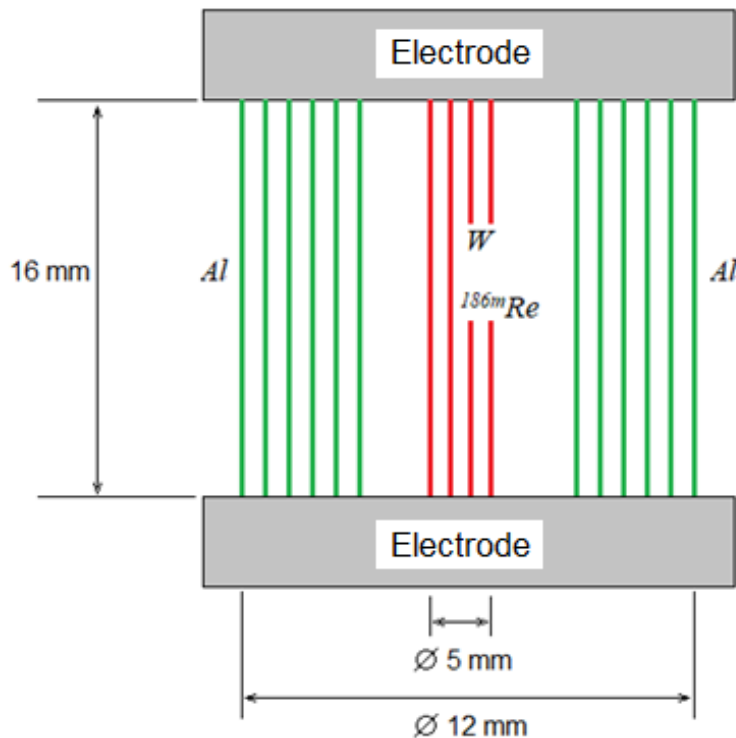
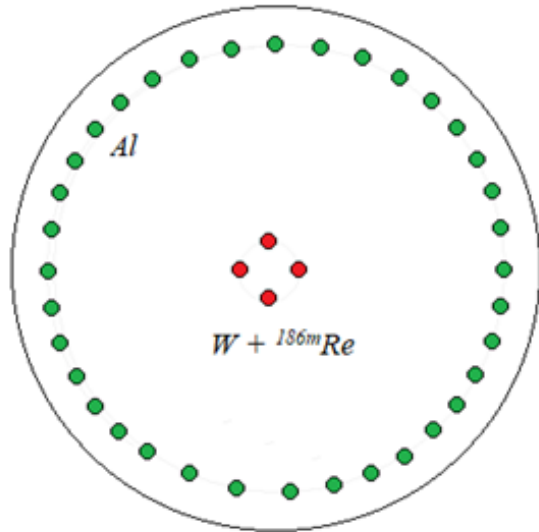
Scheme of EICP facility.

The Angara-5 facility



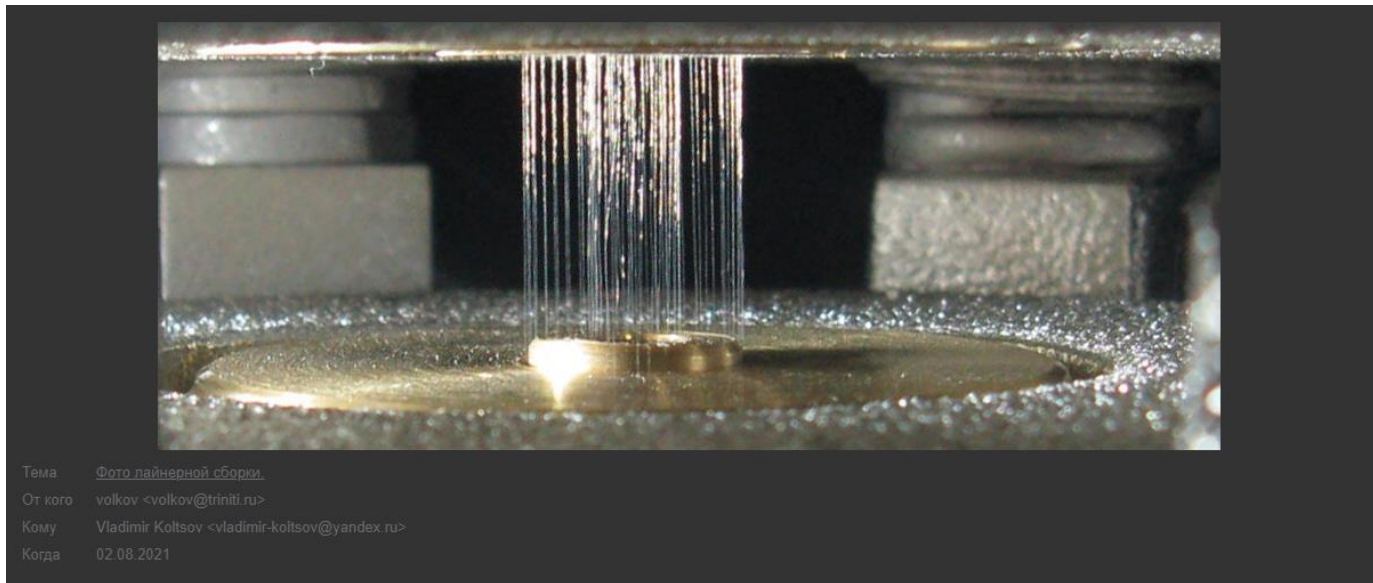
Troitsk Institute of Innovative and Thermonuclear Research (TRINITI).

Angara-5: The composite liner for plasma formation with ions of ^{186m}Re isomers



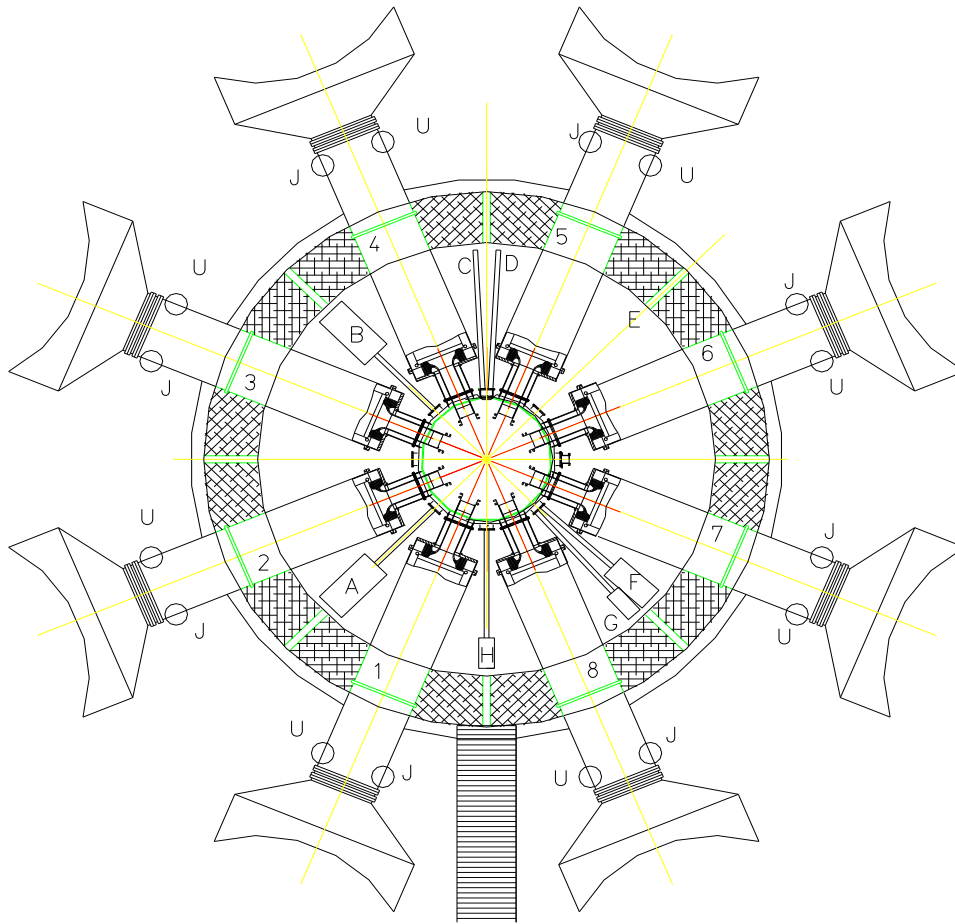
- A composite liner is necessary because it is impossible to strongly heat a plasma made only of heavy ions due to the high intensity of their radiation (Volkov, Zaitsev, Grabovsky, ..., 2010 [19]).
- During a high-current electrical discharge, forty Al wires $\varnothing 18\ \mu\text{m}$ of the outer cylindrical layer form a high-temperature dense compressed Z-pinch plasma.
- The material of four $W + ^{186m}\text{Re}$ wires $\varnothing 6\ \mu\text{m}$ inside the cylinder also transforms into high-temperature plasma.

Composite liner for plasma formation with ^{186m}Re ions



- During an electrical implosion of the liner, the flying plasma matter is collected on stainless steel collectors.
- Then the plasma matter is washed off from the collectors, dried, and the γ -spectrum of this sample is measured on a γ -spectrometer.
- The degree of disturbed radioactive equilibrium determines the probability of deexcitation of the isomer in the plasma.

Plasma diagnostic devices of the Angara-5 facility



The shaded annular area is a concrete protection with an internal diameter of 8.5m.

1 - 8 – module numbers.

Plasma diagnostics were carried out using X-ray spectroscopy, measuring total radiation losses and radiation power with a thermocouple calorimeter, and the vacuum X-ray diodes (VXD) technique with filters. The dimensions of the emitting region were determined by a pinhole camera and multi-frame recording of X-ray images.

A – camera obscura.

B – CXP6 recorder.

C – thermocouple calorimeter.

D – vacuum X-ray diodes (VXD) system .

E – output of radiation to optical scanning.

F – technique for measuring the characteristic L-radiation of tungsten.

G – crystal X-ray spectrograph.

H – spectrometer with a reflective diffraction grating.

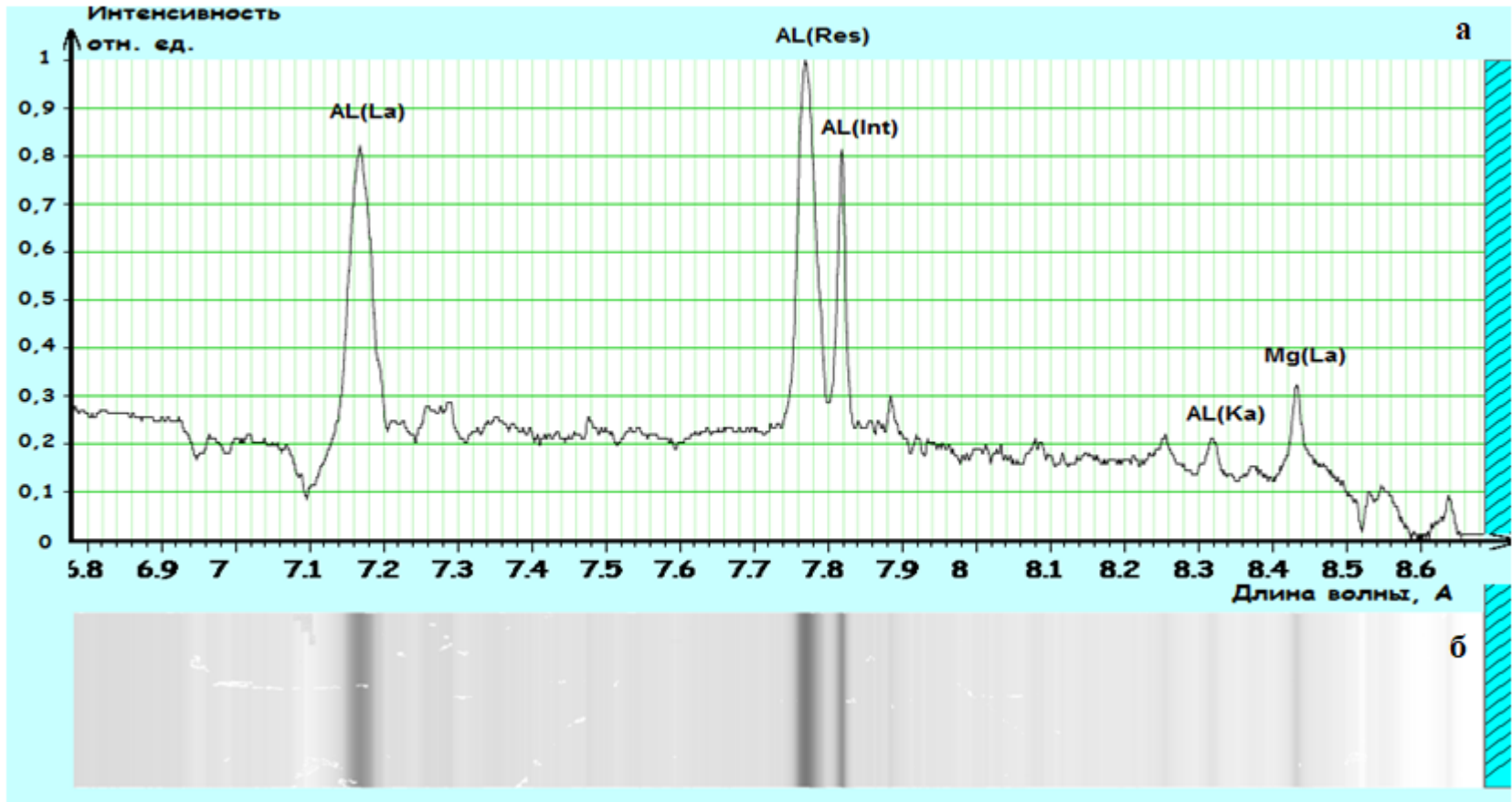
J – current derivative sensor at the beginning of the water transmission line.

U – voltage sensor at the beginning of the water transmission line.

Laser sensing - direction between

6-7 and 2-3 modules.

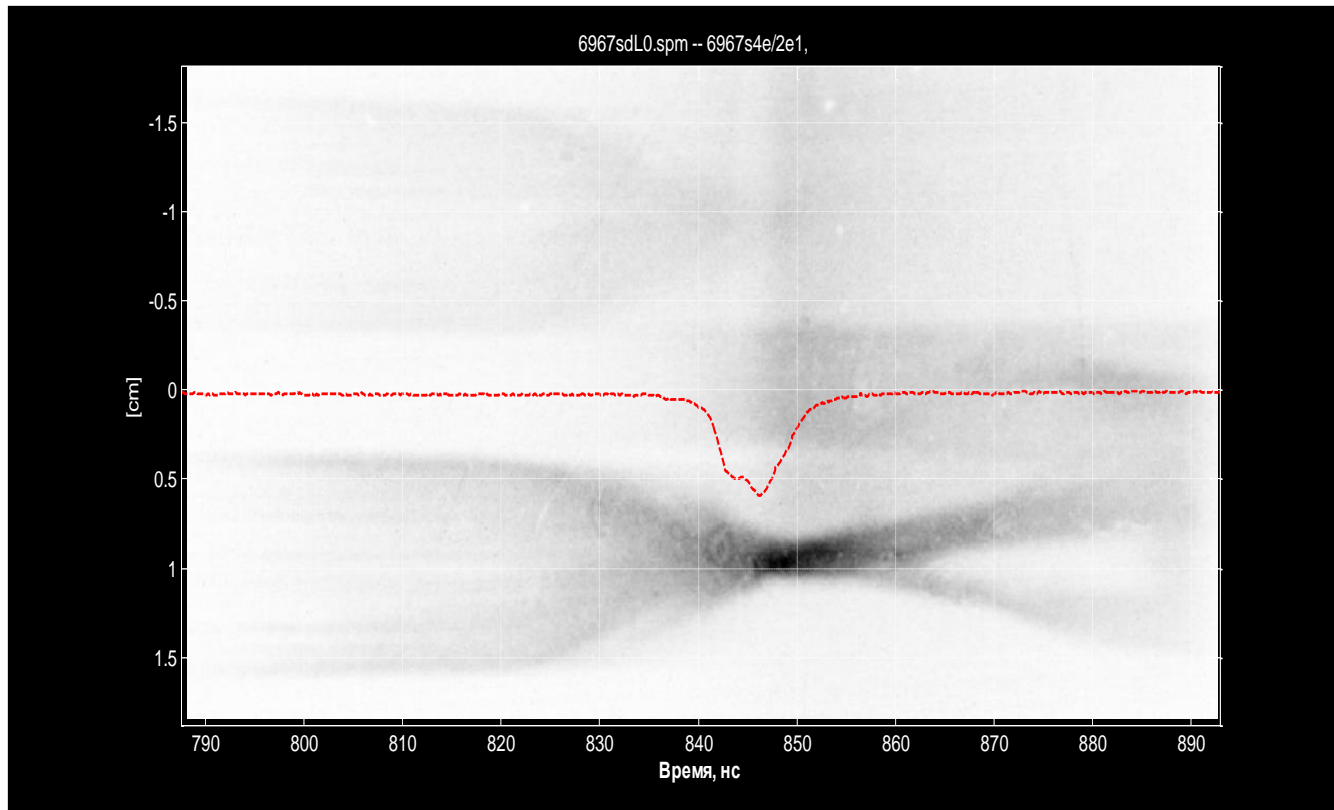
Determination of plasma temperature from the X-ray spectrum



- a) digitized image of the spectrum from X-ray film, taking into account the transmission of filters and the integral reflection coefficients of mica crystals in the second order of diffraction.
- b) Spectrum on X-ray film obtained with a crystal focusing spectrograph.

The electron temperature and plasma density were determined from the ratio of the intensities of spectral lines based on the collisional-radiative plasma model.

Optical diagnostics of Angara-5 plasma



- Radial optical slot scan of the pinch obtained using the SFER-2 electron-optical recorder. A synchronized X-ray signal $h\nu > 700$ eV is superimposed on the optical scan.
- At a low concentration of heavy metals in the pinch plasma, a temperature of ~ 1 keV is reached, as in the laser plasma of the Iskra-5 facility. Then, due to the longer plasma lifetime, the probability of SDENI will be greater than 10^{-7} in laser plasma, and will be no less than 10^{-6} .

Deposition of the ^{186m}Re isomer to W-wires



W-wire \varnothing 6 μm and Cu-wire \varnothing 1 mm.

Requirements:

- Re-metal in the middle of the W-wire.
- The length of the W-wire is 20 cm using liner assembly technology.

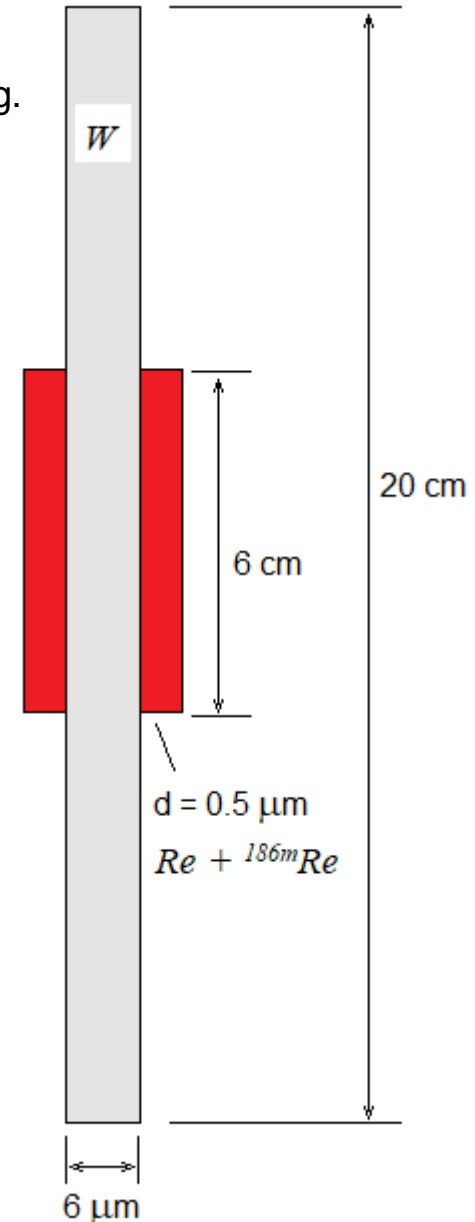
Re deposition method

- Electrodeposition of Re on W-wires.
- Annealing of samples in hydrogen atmosphere to reduce Re to metal.

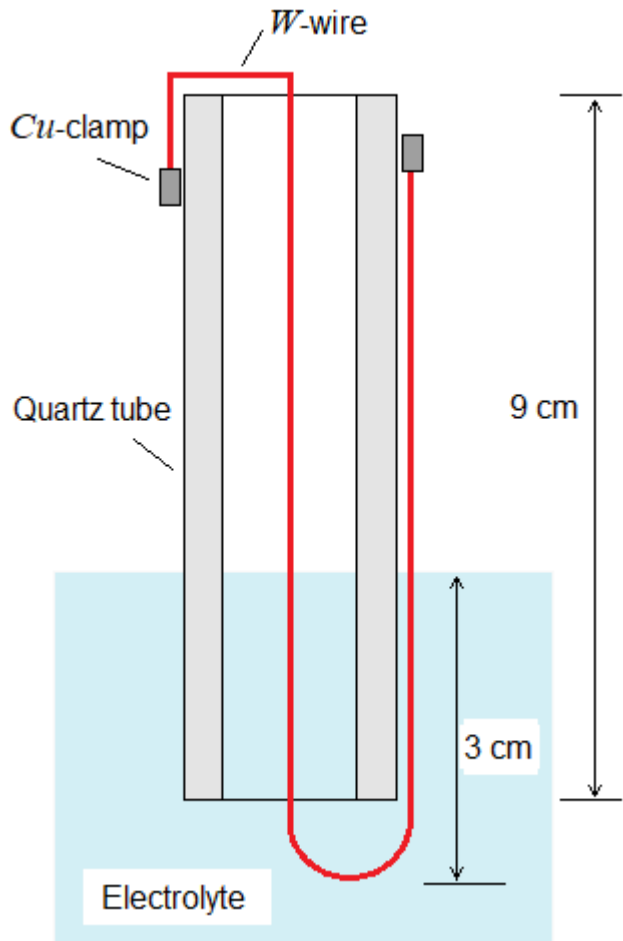
W-wire
for the liner assembling.

In Re-metal
isomer concentration
0.001%.

^{186m}Re activity
0.13 Bq.



Electrodeposition of Re onto W-wires



Electro-deposition:

- Electrolyte 35 ml:
 - Sulfuric acid 70 g/L
 - Ammonium sulfate 40 g/L
 - Ammonium perrhenate NH_4ReO_4 (Re -metal 80 mg)
 - ^{186m}Re isomer 0.001% concentration in Re -metal.
- Electrodeposition onto five cathode modules at once: current ≈ 0.7 mA.
- Control of the mass of precipitated Re by the activity of the ^{186m}Re isomer.

Annealing in a hydrogen atmosphere of Re on W-wires directly in the assembly with quartz tubes.

Cathode module in electrolyte:

W-wire fixed on quartz tube

Time decay of the intensity of the 137 and 40 keV γ -lines from a plasma matter sample.

Experimental conditions:

- Z-pinch plasma with ^{186m}Re isomeric nuclei.
- Plasma contains 10% heavy metals.
- Average $\Theta_e \approx 400$ eV, $\tau \approx 12$ ns, $\eta \approx 0.001\%$.
- Analysis of the plasma matter of 3 plasma shots.
- $P_{\text{SDENI}} \geq 2 \cdot 10^{-7}$.

Sample preparation:

- Washing the plasma matter from the collectors (stainless steel foil) with 500 ml of 5M nitric acid.
- Evaporation and transfer of the plasma matter into a cuvette of HFG detector with a well.
- Activity $^{186m}\text{Re} \sim 0.1$ Bq

1. Clarification of the mechanism of deexcitation of nuclear isomers in plasma:

- Experimental study of the dependence of isomers deexcitation on the electron and ion temperature of the plasma:

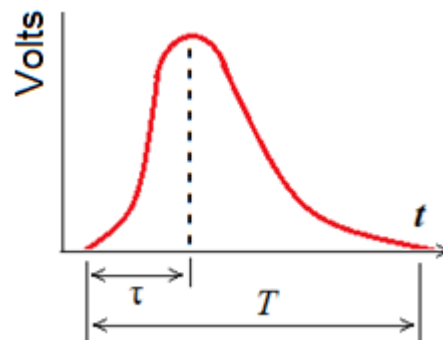
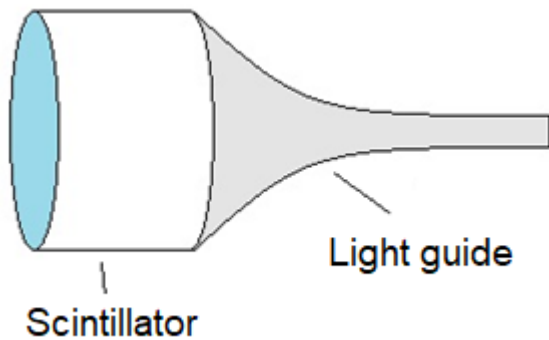
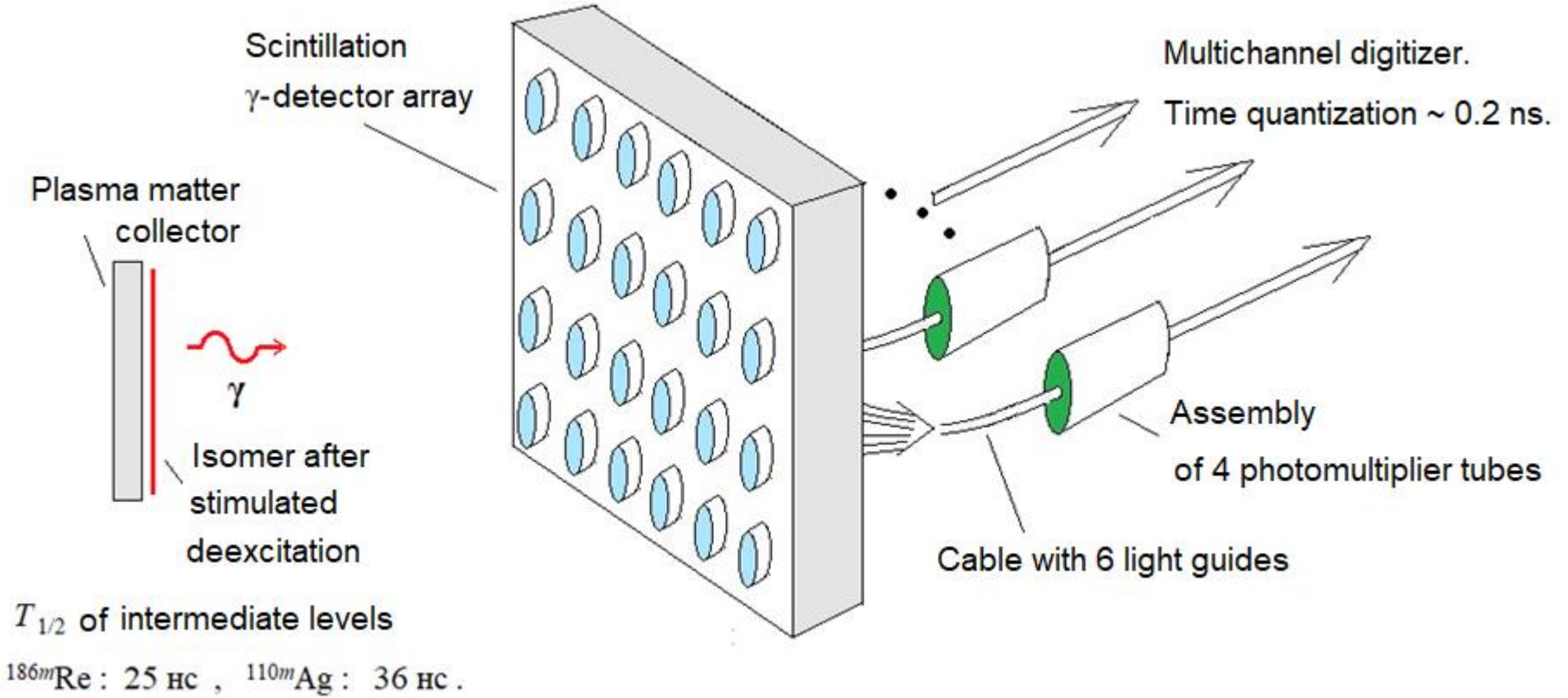
If there are 5% heavy metals, then the average $\Theta_e > 500$ eV, $\tau > 10$ ns, $\Theta_e > 1$ keV for more than 2% of the plasma mass.

- Theoretical and experimental search of possible trigger levels for the ^{186m}Re isomer.
- Study of the influence of plasma irradiation with photons resonant to the nuclear trigger transition.

2. Increasing the sensitivity of the experiment:

- Obtaining material with a higher concentration of isomeric nuclei.
- Development of more sensitive methods for measuring isomers deexcitation (in particular, direct measurement of γ -quanta of stimulated deexcitation of isomers in plasma).

Detection of γ -quanta of the isomers stimulated deexcitation will make it possible to increase the sensitivity of measurements by an order of magnitude (Koltsov, ..., 2017 [21])



Scintillators:

- $\text{LaBr}_3(\text{Ce})$: $\tau = 6$ ns, $T = 20$ ns, energy resolution $\approx 7\%$ for $\gamma = 120$ keV.
- Plastic: $\tau \approx 0.2$ ns, $T \approx 2$ ns.

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Thank you for your attention!