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Computational and theoretical analysis of experiments on irradiation of ^{186m}Re isomer preparations on the Iskra-5 laser facility

D.E. Larin

Abstract. **We describe the formulation and results of experiments obtained using the Iskra-5 facility to induce stimulated de-excitation of the isomeric state of ^{186}Re nuclei by high-intensity direct irradiation, in which a violation of the radioactive equilibrium between the ground and isomeric states is recorded. A comparative analysis of the formulations of two series of experiments is performed. Possible mechanisms for stimulating intranuclear transitions with numerical estimates of the process cross sections are considered. A hypothesis on the stimulation of these transitions by epithermal energy electrons is proposed and possible experiments for its verification are described.**

Keywords: *isomeric states of atomic nuclei, laser plasma, intranuclear transitions.*

1. Introduction

Metastable excited states of atomic nuclei (isomeric states) are characterised by high values of the specific energy stored in a substance consisting of such nuclei. For 12 known isomers with a half-life of more than 10 years, the specific energy ranges from 5 (^{121m}Sn) to 1320 MJ g^{-1} (^{178m2}Hf). In terms of energy intensity, isomeric energy sources could occupy an intermediate position between chemical and nuclear sources if there were methods for stimulated de-excitation of isomeric nuclei with a positive energy balance.

One of the methods for stimulated de-excitation of isomers, which is widely studied both theoretically and experimentally, is the de-excitation of an isomer in a hot dense plasma, in particular when targets containing isomeric nuclei are irradiated with high-intensity laser radiation. A number of authors indicate (see, for example, [1]) that the gamma decay rate increases by several orders of magnitude when the isomers are heated to temperatures comparable to the distances between nuclear energy levels.

In conditions of a hot dense plasma, interlevel transitions in ion nuclei can occur through several channels. In addition to direct interaction with thermal radiation of plasma, nuclear transitions can be stimulated by resonant perturbations of the Coulomb field inside the ion, which occur during bound–bound electronic transitions [nuclear excitation by electronic transition (NEET)] and free–bound electronic transitions [nuclear excitation by electronic capture (NEEC)] initiated, in particular, by inelastic collisions with the hot electron component of the plasma. The excitation of a nucleus from the isomeric level to the overlying one by the NEEC mechanism (with subsequent decay bypassing the long-lived isomeric level) was experimentally confirmed in work [2] by measuring the intensity of gamma decay of ^{93m}Mo nuclei upon recombination of ions containing these nuclei.

One of the isomers promising for the development of an energy source controlled in a wide power range is the isomeric state of the ^{186}Re nucleus, the energy level diagram of which is shown in Fig. 1 (see work [3]). The small transition energy of 2.7 keV between the metastable 8^+ level with a half-life of 2×10^5 years and the short-lived 3^- level makes this nucleus attractive for use in experiments on stimulated de-excitation in laser plasma.

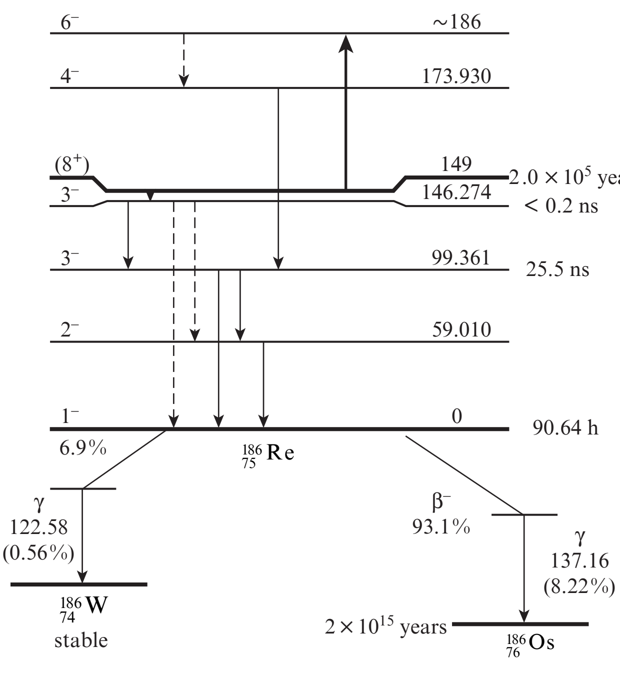

Figure 1. Energy level diagram of the ^{186}Re nucleus.

2. Joint experiments at the V.G. Khlopin Radium Institute and the RFNC-VNIIEF

Experiments on the stimulated de-excitation of ^{186m}Re nuclei were carried out on the Iskra-5 laser facility [4] at the FSUE

D.E. Larin Federal State Unitary Enterprise 'Russian Federal Nuclear Centre – All-Russian Research Institute of Experimental Physics', prosp. Mira 37, 607188 Sarov, Nizhny Novgorod region, Russia,
e-mail: delarin@vniief.ru

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'RFNC-VNIIEF' (Sarov) together with a group of researchers from the V.G. Khlopin Radium Institute (St. Petersburg) [5]. The laser targets manufactured using the V.G. Khlopin Radium Institute technology were steel and tungsten substrates, on which ammonium perrhenate NH_4ReO_4 was applied either by electrolytic deposition (for steel) or by instillation with subsequent drying (for tungsten); then the targets were annealed in a hydrogen atmosphere. The activity of each of the targets was approximately 0.5 Bq. The metal film of rhenium on the substrate had a thickness varying in the range of 1 – 1.5 μm, a diameter of 3 mm, and a rather inhomogeneous distribution.

Laser radiation with a wavelength of 1.312 μm (first harmonic) was directed straight to the sample located at the bottom of a cylindrical duralumin collector. The inner walls of the collector were lined with polyethylene film, on which rhenium evaporated from the target was deposited. One aperture in the collector cover served to enter the laser beam; through the second aperture, the laser plasma radiation fell into the pinhole camera with an X-ray photographic film, which was used to determine the laser spot size and the irradiation intensity. The target splinters were extracted from the collector and subjected to spectrometric analysis in order to establish the violation of the radioactive equilibrium between the ground ^{186g}Re and the excited ^{186m}Re states of rhenium-186 nuclei. The laser spot diameter on the target surface was 600 μm, the FWHM duration was 0.3 ns, the pulse energy was 300 J, and the corresponding intensity of laser radiation on the target surface was 3×10^{14} W cm^{-2}. The authors of work [5] estimate the fluence of X-ray photons with energies near 3 keV as 10^{15} cm^{-2} keV^{-1}; other laser plasma parameters were not evaluated.

The spectrometric analysis of the target splinters was performed at the V.G. Khlopin Radium Institute. As a result of one of the experiments, noticeable temporal fluctuations in the intensity of the gamma-ray decay line of the ^{186g}Re state were detected. This dependence is shown in Fig. 2; the error is given at a level of 1σ; the exposure of each measurement was 12 hours.

Based on the results of a series of experiments, the authors of work [5] conclude that the transition of the ^{186}Re nucleus from the m state to an unknown level lying below the metastable one and having a half-life of about 10 days has been successfully stimulated.

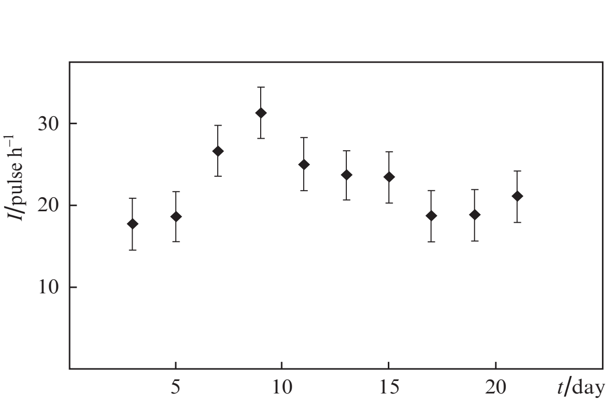

Figure 2. Time dependence of the ^{186g}Re decay rate (spectrometric measurements at the V.G. Khlopin Radium Institute).

3. RFNC-VNIIEF experiments

To reduce the statistical error of the result obtained, as well as due to the impossibility of carrying out measurements at the V.G. Khlopin Radium Institute in the first two days after irradiation associated with the transportation of splinters, a second series of experiments on stimulated deexcitation of ^{186m}Re nuclei in a modified formulation was conducted on the Iskra-5 facility.

The targets were made of titanium substrates with a thickness of 2 mm, in which 'wells' were drilled with a diameter of 1.3 mm and a depth of 1.8 mm. A solution of ammonium perrhenate was poured into the wells, which after drying formed a solid array of salt; the activity of each target was approximately 30 Bq. The targets were not annealed. The substrates with salt-filled wells were placed in the same duralumin collector as in the first series of experiments.

The result with the most pronounced effect from the entire series is described in work [6]. In this experiment, the laser pulse duration was 0.3 ns, the pulse energy was 270 J, and the effective laser spot diameter was either 100 μm (50% of the laser radiation energy is concentrated in the spot) or 300 μm (90% of the laser energy is concentrated in the spot). The corresponding laser intensities were equal to 10^{16} or 1.3×10^{15} W cm^{-2}; thus, the intensity of the incident laser radiation in the brightest region of the spot was two orders of magnitude higher than in the first series of experiments.

The simulation of the experiment was performed using the MIMOSA calculation complex [7] in a two-dimensional statement in the approximation of local thermodynamic equilibrium and uniform laser spot distribution with an intensity of 10^{15} W cm^{-2}. The calculation has shown that the maximum values of the electron, ion and photon temperatures in the laser plasma formed by an isomeric preparation at the laser spot centre by the time moment of 0.37 ns reach 2.0 keV, 150 and 100 eV, respectively.

In the process of spectrometric measurements of target splinters, the detector sensitivity was controlled by the gamma line intensities of the detector contamination, thus used as benchmarks. Within the limits of the statistical measurement error, no deviations from the exponential law of contamination decay were detected.

Figure 3 shows the intensity dependence of the decay gamma lines of the ^{186}Re ground state. The error is indicated

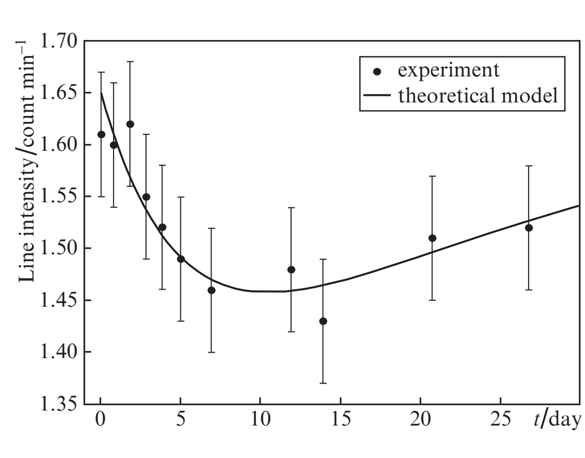

Figure 3. Experimentally obtained and calculated dependences of the ^{186g}Re decay intensity on time (spectrometric measurements at the RFNC-VNIIEF).

at a level of 1 s; the exposure of each measurement was 19 hours. The presented experimental dependence is analysed in detail in work [6]. The analysis allowed the authors (in contrast to the assumptions of work [5]) to put forward a hypothesis on the transition of a fraction of the ^{186m}Re nuclei to a level with an energy higher than that of the ^{186m}Re state. According to the estimate, the half-life of the hypothetical level is 400 h, its energy amounts to $E = 3.2$ keV relative to the ^{186m}Re state, and the spin-parity is 7^+. The fraction of ^{186m}Re nuclei that experienced the $8^+ \rightarrow 7^+$ transition to the hypothetical level was 10%; more correct data processing carried out later gave a value of 18%. These parameters were used to estimate the cross section of the transition process.

4. Analysis of experimental results

When considering the possible mechanisms for stimulating the transitions in the ^{186}Re nucleus, we should note, first and foremost, that the probability of a transition directly under the action of laser radiation, estimated in work [6] in the semiclassical approximation, is negligible, that is, on the order of 10^{-21}. If we consider the stimulation by X-ray quanta, then, by definition, the process cross section is

$$\sigma = \frac{\mathrm{d}N}{\Phi}, \tag{1}$$

where dN is the fraction of nuclei that experienced the transition, and Φ is the fluence of stimulating quanta. The calculated spectrum of X-ray radiation in the target volume, integrated over the plasma lifetime, is shown in Fig. 4 (see work [6]). It can be seen that $\Phi = 15$ J keV^{-1} in the region occupied by the laser radiation spot, which, taking into account the spectral group width of 15 eV, gives the number of quanta 4.7×10^{14}. The calculation of the X-ray radiation spectrum was performed for a laser radiation intensity of 10^{15} W cm^{-2}, which corresponds to its average intensity in the region with a diameter of 300 μm. Then the cross section of the $8^+ \rightarrow 7^+$ transition is

$$\sigma_E = \frac{\mathrm{d}N}{\Phi_E} = \frac{18\%}{15\dfrac{\mathrm{J}}{\mathrm{keV}}\,\dfrac{1}{3.2\ \mathrm{keV}}} \times$$

$$\times\, \pi(150\ \mu\mathrm{m})^2 = 4.4 \times 10^3\ \text{barn keV}. \tag{2}$$

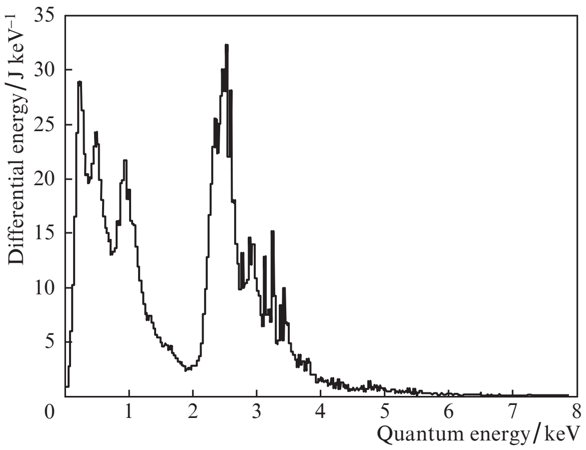

Figure 4. Spectrum of X-ray quanta propagating deep into the target (the group width is 15 eV).

On the other hand, according to the Breit–Wigner formula, the resonant process cross section at the transition energy $E \gg \Gamma$ is

$$\sigma_E \approx \pi\left(\frac{\hbar c}{E}\right)^2 \frac{2J_\mathrm{f}+1}{2J_\mathrm{i}+1}\Gamma = 5 \times 10^{-17}\ \text{barn keV}, \tag{3}$$

where Γ is the sum of the level widths, and J_i and J_f are the moments of the initial and final energy levels, which implies that the process cross section is the same 20 orders of magnitude smaller than the estimates obtained from the results of the experiment.

Thus, the fluence of X-ray quanta in the line is

$$\Phi_E = \frac{15\dfrac{\mathrm{J}}{\mathrm{keV}} \cdot \dfrac{1}{3.2\ \mathrm{keV}}}{\pi(150\ \mu\mathrm{m})^2} \sim 10^{19}\ \mathrm{keV}^{-1}\ \mathrm{cm}^{-2}. \tag{4}$$

This means that the fluence of quanta of the bremsstrahlung spectrum with energies near 3 keV, claimed in work [5], equal to 4×10^{17} keV^{-1} cm^{-2}, is insufficient to detect an effect exceeding the statistical measurement error.

The actual value of the X-ray fluence may turn out significantly lower, since the MIMOSA models do not take into account the processes of generation and transfer of electrons with epithermal energies. The fraction η of laser radiation energy converted into the energy of hot electrons in the case of high laser radiation intensities is approximately described by the formula [8]:

$$\lg\eta = 1.3\lg\left(\frac{I\lambda^2}{10^{15}}\right) - \left\{\left[1.3\lg\left(\frac{I\lambda^2}{10^{15}}\right)\right]^{3/2} + 1\right\}^{2/3}, \tag{5}$$

where I is the peak intensity of laser radiation (at 10^{14} W cm^{-2}), and λ is the laser wavelength equal to 1.312 μm (the first harmonic of the Iskra-5 laser facility).

Substituting the corresponding values in formula (5), we find that the energy fraction of hot electrons in the total energy balance is 30%. For comparison, in the first series experiments performed with the participation of the V.G. Khlopin Radium Institute, the fraction of hot electrons was 7%.

Thus, the estimate for the $8^+ \rightarrow 7^+$ transition cross section, under assumption that the entire energy of electrons with epithermal energies is resonantly spent on the nucleus transition, is

$$\sigma = \frac{\mathrm{d}N}{\Phi_\mathrm{el}} = \frac{\mathrm{d}N\,S}{\eta E_\mathrm{las} E^{-1}} =$$

$$= \frac{0.18\pi(50\ \mu\mathrm{m})^2}{30\% \cdot 270\ \mathrm{J} \cdot 90\% \cdot (3\ \mathrm{keV})^{-1}} \approx 90\ \text{barn}, \tag{6}$$

where Φ_el is the electron fluence, S is the laser spot area, and E_las is the laser pulse energy. Substituting the values specified in work [5] and the proportion of hot electrons into formula (6), we obtain $\sigma = 10^{-7}\,\pi(300\ \mu\mathrm{m})^2\,[7\%\ 300\ \mathrm{J} \times (3\ \mathrm{keV})^{-1}]^{-1} \approx$ 6 mbarn, which is 4 orders of magnitude less than in formula (6).

In this case, either the estimate of the relative number of nuclei that have passed to the hypothetical level, which, according to the results of experiments at the V.G. Khlopin

Radium Institute, is 10^{-7}, is greatly underestimated, or, what seems most likely, due to the small thickness of the rhenium target, the electrons spent only a small fraction of their energy on inelastic interactions with ^{186m}Re nuclei. More precise conclusions can be drawn from the results of computer simulation of the transfer of electrons of the corresponding spectra in the laser targets.

5. Discussion of the results

The results of two series of experiments in different formulations, aimed at stimulated de-excitation of the ^{186m}Re isomer under direct irradiation with a laser pulse on the Iskra-5 facility have been compared. Based on the results of the analysis of spectroscopic measurements of the irradiated preparation, we have made an assumption on the significant role of hot (having epithermal energies) laser plasma electrons in the interactions of the nucleus with the electron shell of ions, which leads to transitions between energy levels in the nucleus. Estimates of the laser energy spent on the generation of hot electrons and fluences of hot electrons, as well as upper estimates of the stimulated transition cross section to the assumed 7^+ level in the ^{186}Re nucleus, are obtained.

A comparative analysis of the experiments of both series shows that the conditions developing in the laser plasma differ significantly between them in terms of the fluence of hot electrons. If we assume that hot electrons play a decisive role in stimulating intranuclear transitions, then the geometry and manufacturing technology of targets become of great importance for the transport of electrons, their deceleration in matter and the processes of inelastic interaction with atoms. In this case, the fundamental differences between the results obtained in the experiments of both series are explained. It should be noted that neither the flux nor the spectrum of hot electrons was measured in both series of experiments. Therefore, in the formulation of experiments to verify this hypothesis, appropriate laser plasma diagnostic systems should be provided. An obvious method for experimental verification of the proposed hypothesis on the effect of electrons with epithermal energies on the internal state of the atomic nucleus is the irradiation of thin targets containing the ^{186m}Re isomer with monochromatic electrons of different energies (for example, in the range 0–10 keV). However, in such an experiment, the irradiated target will be in a condensed state, which makes it difficult to assess the effect of the thermodynamic state of the preparation on the intensity of the processes of intranuclear transitions.

Setting up an experiment on any other laser facility is complicated by the fact that to achieve a significant effect, not only a high laser radiation intensity I is required, but also a high pulse energy E, since the number of hot electrons is approximately $N_{\rm el} \propto E \lg I$.

In this regard, it seems appropriate to perform an experiment on the irradiation of preparations of the ^{186m}Re isomer with laser radiation in a solution containing metal nanoparticles, similar to the experiments described in work [9], during which the effect of laser radiation on the gamma activity of ^{152}Eu was established. This isotope is similar in structure to ^{186}Re, that is an odd–odd nucleus with an isomeric level of ^{152m}Eu with an energy of 45.6 keV and a lifetime of 13.434 h, which allows us to expect changes in the decay rate of both the g and m state of the ^{186}Re nucleus.

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