Laser-induced synthesis and decay of Tritium under exposure of solid targets in heavy water

E.V. Barmina[1], S.F. Timashev[2,3], and G.A. Shafeev[1,3]

[1] Wave Research Center of A.M. Prokhorov General Physics Institute of the Russian Academy of Sciences, Moscow, Russian Federation

[2] L.Ya. Karpov Institute of Physical Chemistry, Moscow, Russian Federation

[3] National Research Nuclear University MEPhI, Moscow, Russian Federation

Abstract

The processes of laser-assisted synthesis of Tritium nuclei and their laser-induced decay in cold plasma in the vicinity of solid targets (Au, Ti, Se, etc.) immersed into heavy water are experimentally realized at peak laser intensity of 10^{10}-10^{13} W/cm^2. Initial stages of Tritium synthesis and their laser-induced beta-decay are interpreted on the basis of non-elastic interaction of plasma electrons having kinetic energy of 5-10 eV with nuclei of Deuterium and Tritium, respectively.

PACS: 42.62.–b; 61.46.+w; 78.66.–w

Recent theoretical work shows the capability of laser radiation to directly excite nuclear levels of energy [1]. However, exciting a nuclear transition would require an X-ray or gamma-ray lasers with an intensity greater than 10^{20} Watts/cm^2. At the same time, laser generated photons can indirectly influence the nuclear levels and alter or induce nuclear reactions. Laser beams at intensity level of 10^{18} W/cm^2 are capable of inducing nuclear transformations under laser exposure of solid targets in vacuum. This can be achieved at pico- and femtosecond laser pulse durations [2-4]. The yield of 10^3 alpha particles per pulse has been detected at peak laser intensity on the target of 2×10^{18} W/cm^2 [5]. In accordance with [5], the mechanism of initiation of a neutron-less fusion reaction is due to the acceleration of electrons and protons in the plasma separated by strong laser field.

New possibilities of laser initiation of nuclear reactions have been demonstrated at peak laser intensity levels of 10^{10}-10^{13} W/cm^2 [6-9]. The crucial role in laser-induced acceleration of nuclear decays belongs to nanoparticles (NPs) and nanostructures (NS) on the target immersed into liquid that interact with laser beam in presence of unstable isotopes. In fact, the possibility of initiation of nuclear transformations in cold laser-induced plasma with electron temperature of 5-10 eV has been shown. Relatively low laser intensity of 10^{12}-10^{13} W/cm^2 can be enhanced both on NPs and NS by a factor of 10^6-10^8 due to plasmon resonance of charge carriers in the nano-entities presented in the solution. In this way the electric field of laser radiation becomes comparable to intra-atomic field and may cause the distortion of electronic shells.

Tritium is another unstable isotope with half-life of 12.2 years. It would be interesting to induce its beta-decay by exposure to laser radiation, as it was done previously for other nuclides. In this work we demonstrate for the first time the nuclear synthesis and decay of Tritium under laser exposure of various targets in D_2O [15].

Different targets were used for exposure of bulk metal targets in a heavy water with some tritium content (D_2O + ξDOT), where ξ stands for molar fraction of D_2O with T. The content of T was measured according to its beta-activity with the use of a Perkin-Elmer beta-spectrometer Tri-Carb 3110TR Liquid Scintillation Analyzer with fluorescent dye ULTIMA GOLD^{TM} cocktail. The accuracy of measurements was 1%. Deuterium content was 98.8 %. Two laser sources were used for exposure of various targets in heavy water. The first source was a Cu vapor laser, pulse duration of 15 ns, wavelength of 510.6 nm, and repetition rate of 15 kHz. The estimated peak power on the target was 10^{10} W/cm^2. The second laser source was a Nd:YAG laser Duetto^{TM}, Timebandwidth, with pulse duration of 10 ps and repetition rate of 50 kHz. Three harmonics of this laser, first at 1064 nm, second at 532 nm, and third at 355 nm were used for target exposure. The estimated peak power on the target was 10^{13} W/cm^2, 5×10^{11} W/cm^2, and 2×10^{11} W/cm^2 at 1064, 532, and 355 nm, respectively.

Laser exposure of various targets placed into a glass cell with liquid was carried out by focusing laser beam on the target with spherical lens of 5 cm focal distance. Laser beam was focused on the target from bottom through a window made of fused silica to a spot on the target of 50 μm in diameter. The targets had the shape of rods with diameter about 1 mm. The sides of the targets were electrically isolated from the solution. Typically, 2 ml of heavy water was placed inside a glass cell cooled by flowing water. The inner diameter of the glass cell was 1 cm. In another set of experiments the metallic target was cathodically biased with respect to another electrode made of Platinum wire 0.5 mm in diameter. The applied voltage was 25 V DC. To ensure the conductivity of the solution, 15 mg of metallic Na were added to 5 ml of D_2O thus producing conducting solution of NaOD.

In the first set of experiments the ablation of targets was carried out in 99.9% pure D_2O (supplied by Euriso-top) without Tritium content. In this case the content of DOT molecules in D_2O was at the background level of the beta-spectrometer, which corresponds to $\xi = 3.54\cdot10^{-14}$. In one set of experiments the voltage was not applied to the target, in the second set of experiments the ablation was carried out simultaneously with electrolysis. Application of voltage to electrodes is accompanied by reduction of Deuterium (and Tritium) on the target as bubbles. Isolation of the sides of target from the solution confined the emission of reduced gas to ablated area.

Typical exposure time of the targets was 1 hour. The target material is dispersed into surrounding liquid in the shape of nanoparticles. A deep crater is formed in the target surface after laser exposure. The inner surface of the crater and the target surface around it are decorated with dense array (4×10^9 cm^{-2}) of nanostructures with average lateral size of 100 nm (Fig. 1). Nanoparticles ablated from the target form a colloidal solution. Their presence in the beam path hinders the exposure of the target especially at the wavelength of the third harmonics (355 nm) due to strong absorption of laser radiation.

Table 1. Laser exposure of targets in pure D_2O with addition of NaOD ($\xi = 3.54 \cdot 10^{-14}$).

№	Material	Wavelength of laser beam, nm	Intensity of laser radiation, W/cm^2 and pulse width	Time of irradiation, min	Electrolysis	Activity, Ci/l	Change with respect to initial level
1	Initial D_2O	-	-	-	-	8.64×10^{-8} $\xi=3.54\cdot10^{-14}$	-
2	Au	532	7×10^{11} 10 ps	60	+	4.06×10^{-4} $\xi=1.66\cdot10^{-10}$	Increase 4.7×10^3
3	Au	1064	10^{13} 10 ps	32	-	1.62×10^{-6} $\xi=6.63\cdot10^{-13}$	Increase by factor 18.8
4	Ti	532	7×10^{11}	62	+	4.74×10^{-4} $\xi=1.94\cdot10^{-10}$	Increase 5.5×10^3
5	Ti	1064	10^{13}	90	-	1.57×10^{-6} $\xi=6.42\cdot10^{-13}$	Increase by factor 18.2
6	Ti	355	2×10^{11}	61	+	9.62×10^{-8} $\xi=3.94\times10^{-14}$	Increase by factor 1.11
7	Pd	355	2×10^{11}	75	+	2.46×10^{-6} $\xi=10^{-12}$	Increase by factor 28.2
8	Pd	1064	10^{13}	62	+	2.31×10^{-6} $\xi=9.47\times10^{-13}$	Increase by factor 26.7

It was found that laser exposure of various targets in D_2O leads to formation of Tritium, and its content depends on several experimental parameters. The results are presented in Table 1. Highlighted samples were exposed under cathodic bias. Laser ablation of targets without electrolysis leads to the increase of Tritium activity to the level which is about 20 times above the background (10^{-7} Curie/l). The synthesis of Tritium is even more pronounced if the ablated target is cathodically shifted. Ablation of either Ti or Au targets with electrolysis results in synthesis of Tritium to the level, which is 10^3 times above the background. The largest change of activity is observed at laser wavelength of 1064 nm. The optical density of the solution at the wavelength of the third harmonics of a Nd:YAG laser (355 nm) is high, especially after generation of some amount of nanoparticles, so the intensity of the laser beam on the target is much lower than at 1064 nm.

The following set of experiments was carried out in heavy water with initial Tritium content. Laser ablation of bulk targets for 40-60 min leads to generation of nanoparticles (NPs). In case of Au and Se targets these NPs are made of Au and Se, respectively [16, 17]. Both these colloids absorb in the visible range of spectrum and are reddish in appearance. The solution without NPs (or with small remaining fraction of NPs) was tested for Tritium content about one week after laser exposure. Tritium content in the initial D_2O was also measured in the same set of samples. Then in another set of experiments the generated colloids were exposed to laser radiation in absence of the target. Tritium content increases by a factor of 1.9 after ablation of a bulk Se target with 10 ps pulses of the second harmonics of a Nd:YAG laser. Further laser exposure of Se NPs solution does not alter this increased level of Tritium activity. Again, exposure of the colloidal solution to radiation of a Cu vapor laser leads to decrease of T activity by a factor of 6.7. Qualitatively similar results are obtained for NPs of Au (Table 2).One can see that activity of Tritium (its content) is higher by a factor of 1.8 than in the initial D_2O right after laser generation of Au NPs with picosecond laser pulses. Further laser exposure of the colloidal solution alone does not alter much the activity of T. On the contrary, small decrease of T activity is observed in case of exposure of Se colloidal solution with 15 ns pulses of a Cu vapor laser.

The exposure of different targets (Au, Ag, Be) to radiation of a 15 ns laser pulses has small effect on the activity of Tritium. The exception is exposure of Se target to this radiation that markedly reduces T content in the exposed solution (see Table 2).

Laser exposure of targets that are cathodically biased in D_2O with relatively high DOT content leads to some decrease of Tritium activity. These results are presented in Table 2.

Table 2. Decay of Tritium activity during laser exposure of different targets with electrolysis.

No	Material	Kind of liquid	Wavelength of laser beam, nm	Intensity of laser radiation, W/cm^2	Time of irradiation, min	Activity, Ci/l
1	Initial D_2O+ DOT	D_2O+ DOT	-	-	-	1.22×10^{-3} ($\xi = 0.5\cdot10^{-9}$)
2	Au	NaOD+DOT	532	7×10^{11}	60	1.12×10^{-3}
3	Au	NaOD+DOT	1064	10^{13}	30	9.03×10^{-4}
4	Ti	NaOD+DOT	532	7×10^{11}	60	1.11×10^{-3}
5	Ti	NaOD+DOT	1064	10^{13}	90	3.35×10^{-4}
6	Pd	NaOD+DOT	1064	10^{13}	90	1.22×10^{-3}
7	Ti	$Li_3C_6H_5O_7$+ DOT	1064	10^{13}	54	6.35×10^{-4}

The final value of activity after laser exposure is around 10^{-3} Ci/l. The variation of activity caused by laser ablation is well pronounced even at high level of initial Tritium activity. As it follows from data presented in Table 1, at low initial Tritium content the increase of Tritium content after laser exposure is observed. One may suggest that this result is the competition between laser-induced decay of Tritium and its formation.

The efficiency of nuclear processes occurring in the course of heavy water electrolysis can depend on the character of roughness of the electrode surfaces on a nanometer scale, the “spikiness” parameters [18, 19] in particular. Indeed, it is precisely in the regions of the sharpest surface relief alterations that high electric field strengths making for the acceleration of electrons and high mechanical stresses depressing the activation barriers for electrochemical processes can both get realized. This parameter is out of control in most experiments with electrolysis of heavy water. On the contrary, laser ablation of metallic targets by sub-nanosecond laser pulses leads to formation of self-organized nanostructures (NS) on the target. The average size and density of NS depends on laser fluence on the target and target material. Therefore, laser ablation technique used in the present work provides guaranteed “spikiness” of the target that may generate high electric fields in the vicinity of the ablated target. Local electric field near the tips of NS under cathodic bias can amount to 10^9 V/m, and they may serve as efficient emitters of high energy electrons.

The observed phenomena of T synthesis cannot be ascribed to nuclear fusion in its classical sense. There should be a low-energy channel that may alter the interaction of deuterons. One may suggest the following scheme of reactions [22, 23]. If the original nucleus is stable (for the sake of definiteness, let it be a deuteron d^+) and the kinetic energy E_e of the electron is too low for any nuclear process involving the breakdown of the original nucleus to take place, the electron-nucleus interaction can result in the emission of a neutrino ν (an irreversible process!) and formation in the nucleus of an intermediate vector boson W^- as a virtual particle (the electron cannot directly interact with a quark). Thereafter there forms a d quark, as a result of interaction between the virtual vector boson W^- and one of the u quarks of a nuclear proton, and the latter converts into a virtual neutron. No actual neutron can form in the nucleus. True, if this were the case, the nucleus being formed would decay, because a two-neutron system is unstable [24]. But for such decay into two neutrons to take place, the mass of the nucleus being formed, which is equal to the sum of the masses of the deuteron and electron, is insufficient. (In the case under consideration, the energy deficit ΔQ is $(m_p + m_e - 2m_n)c^2 \approx -3.01$ MeV, where c is the velocity of light in vacuum and m_p, m_e, and m_n are the masses of the proton, electron, and neutron, respectively.) The conclusions drawn in [24] that the existence of a nuclear stable dineutron is impossible does not relate to the nucleus being introduced here with its mass less than the total mass of two neutrons, despite its zero electric charge, baryon number equal to 2, and zero leptonic charge. We will refer to such a nucleus as β-dineutron and represent the sequence of the processes being discussed as follows:

$$e^-_{he} + d^+ \rightarrow {}^2n_{isu} + \tilde{\nu}\,, \qquad (1)$$

$${}^2n_{isu} \rightarrow d^+ + e^- + \tilde{\nu}\,. \qquad (2)$$

In these expressions, the subscript "he" on the notation of the initial electron points to the initiative character of interaction between the electron and deuteron in the formation of the β-dineutron ${}^2n_{isu}$ in the in-shake-up state (the latter being indicated by the subscript "isu"). Inasmuch as the threshold of such an "inelastic scattering" of the electron by the deuteron is solely determined by the doubled rest mass of the neutrino and amounts to less than 1 eV, the electron kinetic energies of $E_e \sim 10$ eV will apparently be adequate for one to observe such processes. This means that the mass of the β-dineutron practically coincides, as noted above, with the mass of the deuterium atom. The Triton in the experimental conditions of present work may be synthesized according to the following equation:

$$d^+ + {}^2n_{isu} \rightarrow t^+ + n + Q(3.25MeV)\,. \qquad (3)$$

In turn, Tritium nucleus formed in the cold laser plasma may also interact with electrons. According to [22, 23], decay of Tritium may be accelerated ("e-catalysis") owing to this interaction and formation of an

intermediate product β-trineutron $^3n_{isu}$:

$$t^+ + e^- \rightarrow {}^3_0 n_{isu} + \nu \rightarrow {}^3_2 He^{2+} + 2e^- + \nu + 2\tilde{\nu} + Q(0.019 MeV), \quad (4)$$

All experimental results presented above can be interpreted on the basis of the processes (1), (3), and (4). Indeed, processes (1)-(3) dominate at low initial Tritium content in D_2O (10^{-7} Curie/l) and presence of electrons having kinetic energy E_e ~ 5-10 eV in cold plasma. As a result, Tritium content increases by 3 orders of magnitude. On the other hand, predominant β-decay of Triton takes place under relatively high initial Tritium beta-activity ~ 10^{-3} Curie/l.

The laser-induced decay of Tritium is a competing process that determines the stationary level of Tritium during laser exposure. Note that significant decrease of Tritium content occurs during 1 hour laser exposure, which is many orders of magnitude shorter than life time of this nuclide.

Thus, laser-assisted synthesis of Tritium from D_2O has been realized. The synthesis proceeds under exposure of various solid targets with laser pulses. There is definite threshold of the intensity and duration of laser pulses, the synthesis takes place only with short (10 ps) laser pulses and peak intensities exceeding 10^{11} W/cm^2. Tritium is synthesized without any cathodic bias on the target though the application of this bias largely increases the rate of its synthesis. The increase of Tritium content requires careful check. In particular, long-term measurements of fluorescence are required to confirm the decrease of generated Tritium with its characteristic half-life of 12.2 years.

References

1. Wong, A. Grigoriu, J. Roslund, T.-S. Ho, and H. Rabitz, *Phys. Rev.* A 84, 053429 (2011).

2. Andreev A.V., Gordienko V.M., Dykhne A.M., Saveliev A.B., Tkalya E.V., 1997, *JETP Lett*. **66** (5) 312.

3. Andreev A.V.; Volkov R.V.; Gordienko V.M.; Dykhne A.M.; Mikheev P.M.; Tkalya E.V.; Chutko O.V.; Shashkov A.A.; 1999, *JETP Lett*. 69 (5) 343.

4. Schwoerer H., Gibbon P., Düsterer S., Behrens R., Ziener C., Reich C., Sauerbrey R. *Phys. Rev. Lett.*, 86 (11), 2317 (2001).

5. V. S. Belyaev, A. P. Matafonov, V. I. Vinogradov, V. P. Krainov, V. S. Lisitsa, A. S. Roussetsky, G. N. Ignatyev, V. P. Andrianov, *Phys. Rev. E* 72 (2005), 026406.

6. A.V. Simakin, G.A. Shafeev, arXiv:0906.4268; A.V. Simakin, G.A. Shafeev, *Physics of Wave Phenomena*, 16 (4) (2008) 268-274; A.V. Simakin, G.A. Shafeev, *Journal of Optoelectronics and Advanced Materials*, (2010) 12(3), 432 – 436.

7. A.V. Simakin, G.A. Shafeev, *Quantum Electronics* 41 (7) 614 – 618 (2011).

8. A.V. Simakin, G.A. Shafeev, *Physics of Wave Phenomena* (2011) 19(1), 30–38.

9. G.A. Shafeev, Laser-induced nuclear decays in Uranium isotopes, in: Uranium: Characteristics, Occurrence and Human Exposure, Editors: Alik Ya. Vasiliev and Mikhail Sidorov, ISBN: 978-1-62081-207-5, Novapublishers Inc, New York, 2012, p.p. 117-153.

10. Volkovich A.G., Govorun A.P., Gulyaev A.A. Zhukov S.V., Kuznetsov V.L., Rukhadze A.A., Steblevsky A.V., Urutskoev L.I., Lebedev Bulletin of the Lebedev Physics Institute, 2002, No 8, 45 – 50.

11. G.A. Shafeev, A.V. Simakin, F. Bozon-Verduraz, M. Robert, *Applied Surface Science* **254** (2007) 1022–1026.

12. Fleishmann M., Pons S. and Hawkins M. Electrochemically induced nuclear fusion of deuterium, *J. Electroanal. Chem.* 1989. V. 261. P. 301-308.

13. Iwamura Y., Itoh T., Gotoh N., Toyoda I. Detection of anomalous elements, X-ray, and excess heat in a D_2-Pd system and its interpretation by the electron-induced nuclear reaction model, *Fusion Technology*, 1998. V. 33. P. 476-492.

14. Takahashi A. Progress in condensed matter nuclear science. In: Condensed Matter Nuclear Science. Proc. 12th Intern. Conf. on Cold Fusion, Yokohamacity, Japan. 27 Nov-2 Dec 2005. World Scientific Publishing Co. Pte. Ltd.: Singapore, 1-25 p.

15. E.V. Barmina, P.G. Kuzmin, S.F. Timashev, G.A. Shafeev, Laser-induced synthesis and decay of Tritium under exposure of solid targets in heavy water, http://arxiv.org/abs/1306.0830.

16. P.V. Kazakevich, A.V. Simakin, V.V. Voronov, and G.A. Shafeev, *Applied Surface Science*, **252** (2006) 4373–4380.

17. P.G. Kuzmin, G.A. Shafeev, V.V. Voronov, R.V. Raspopov, E.A. Arianova, E.N. Trushina, I.V. Gmoshinsky and S.A. Khotimchenko, *Quantum Electronics*, **42**, (2012) 1042.

18. Timashev S.F., Polyakov Yu.S., Lakeev S.G., Misurkin P.I., Danilov A.I. Fundamentals of fluctuation metrology, *Russian Journal Physical Chemistry* A. 2010. V. 84. No 10, 1807-1825.

19. Mirsaidov U., Timashev S.F., Polyakov Yu.S., Misurkin P.I., Polyakov S.V., Musaev I. *Analytical method for parameterizing the random profile components of nanosurfaces imaged by atomic force microscopy* // Analyst. 2011. V. 136. N 3, P. 570-576

20. S. Lau Truong, G. Levi, F. Bozon-Verduraz, A.V. Petrovskaya, A.V. Simakin, G.A. Shafeev, *Applied Surface Science* **254** (2007) 1236–1239.

21. E.V. Barmina, S. Lau Truong, F. Bozon-Verduraz, G. Levi, A.V. Simakin, G.A. Shafeev, , *Quantum Electronics*, 40 (2010) 346-348.

22. Timashev S. F., V. I. Muromtsev V.I., Akovantseva A.A. Nuclear Processes Initiated by Electrons , Russian Journal of Physical Chemistry A, 2013, Vol. 87, No. 6, pp. 1063–1069.

23. Timashev S.F. “Beta-dineutron” as a radioactive element and its possible role in nuclear synthesis processes (in press).

24. Zel’dovich Ya.B., Gol’danskii V.I., Baz’ A.I. Systematics of the lightest nuclei, Physics – Uspekhi. Advances in Physical Sciences. 1965. V. 8. N 2. P. 177–201.