

Effect of laser radiation on aqueous solutions of beta-active nuclides

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Abstract. Experimental data are presented on the effect of pulsed laser irradiation of aqueous ^{234}Th , ^{231}Th and ^{137}Cs solutions on the concentrations of the beta-active nuclides in the solutions. Central to the investigation technique used is that the solutions contain nanoparticles resulting from the interaction of laser radiation with a target placed in the cuvette. The concentration of the radionuclides was evaluated before, during and after laser irradiation from the area under gamma-ray lines corresponding to spontaneous gamma decay. The gamma photon energies of ^{234}Th , ^{231}Th and ^{137}Cs are 92, 186 and 661 keV, respectively. It has been shown that laser irradiation reduces the concentration of the three beta-active nuclides without excess gamma-ray emission in the energy range of their spontaneous decay. This is possible if decay follows another mechanism, activated by interaction of laser radiation with the aqueous solutions of the nuclides in the presence of nanoparticles.

Keywords: laser radiation, beta-active nuclides, gamma-rays, nanoparticles.

The effect of laser radiation of relatively low intensity (10^{10} to 10^{12} W cm $^{-2}$) on the concentration of a number of nuclides has recently been the subject of several experimental studies. At such intensities, the electric and magnetic fields of the laser radiation are several orders of magnitude weaker than intratomic fields. These are comparable to laser fields at intensities of the order of 10^{18} to 10^{20} W cm $^{-2}$. At such intensities, electrons in the laser-induced plasma can acquire energies up to 1 GeV as a result of laser wakefield acceleration [1–3]. Energies of ~ 1 GeV are comparable to nuclear energy levels and in fact lead to nuclear transmutation in experiments in accelerators [4].

In this paper, we examine the effect of laser irradiation on the concentration of three nuclides that undergo β^- decay: caesium-137, thorium-234 and thorium-231. The half-life of ^{137}Cs is 30 yr. It emits an electron and decays to ^{137}Ba . The latter is formed in an excited state and emits a 661-keV gamma

photon. In addition, there is direct beta decay of ^{137}Cs to the ground state of ^{137}Ba , but the contribution of this decay channel is only a few percent [5] (Fig. 1).

Unlike in previous studies [6, 7], in addition to increasing the number of nuclides under investigation we obtained data on the variation of nuclide concentrations in solution during several laser irradiation cycles.

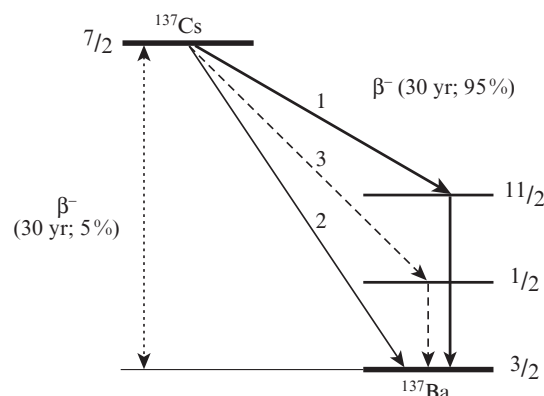


Figure 1. Decay scheme of caesium-137.

Aqueous nuclide solutions were enclosed in a glass cuvette equipped with a cooling water jacket. Laser radiation was incident vertically from below through a glass window. In our experiments with ^{137}Cs , we used a pulsed Ti:sapphire laser with a pulse energy of 2.7 mJ, emission wavelength of 775 nm and pulse repetition rate of 1 kHz. The laser pulse duration was 2.5–3 ps. The laser beam was focused by a lens ($F = 50$ mm) into the bulk of a test solution (2 mL of an aqueous caesium-137 chloride solution). As a laser ablation target intended for nanoparticle generation, we used a NdFeB alloy. In a number of experiments, an unirradiated sample was placed near the gamma detector in order to verify long-term stability of its operation. Nuclide concentration was determined as the area under an appropriate line in the gamma-ray spectrum using application software. The counting time was adjusted so that the area under spectral lines was determined with an accuracy of 0.5% or better.

Figure 2 shows a typical gamma-ray spectrum of an aqueous ^{137}Cs salt solution. This nuclide has one line, at 661 keV. As an unirradiated reference, we used an aqueous ^{134}Cs solution.

^{134}Cs has several gamma-ray peaks, one at 605 keV. It is located away from the 661-keV ^{137}Cs peak and does not distort the area under this peak. In addition, the spectrum contains

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Received 5 April 2017

Kvantovaya Elektronika 47 (7) 627–630 (2017)

Translated by O.M. Tsarev

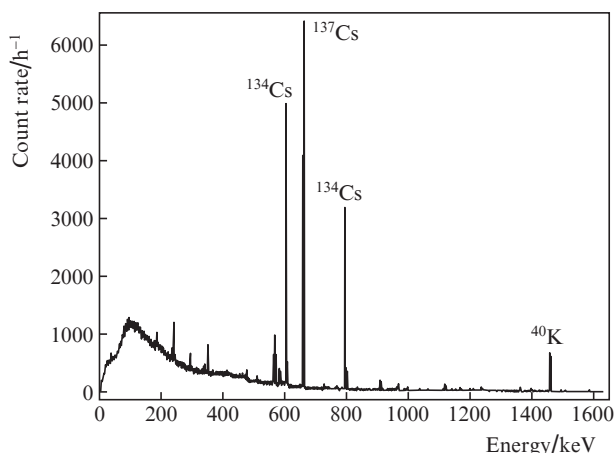


Figure 2. Typical gamma-ray spectrum of a mixture of two caesium isotopes.

a broad low-energy band. This is background gamma radiation, primarily cosmic rays. The intensity of the background fluctuates from day to day, but it is also located far away from the 661-keV peak and introduces no errors into ^{137}Cs activity measurements. Also discernible is a potassium-40 background peak.

Figure 3 illustrates the evolution of ^{137}Cs concentration, measured as indicated above, in a solution exposed to picosecond Ti:sapphire laser pulses. The open circles indicate the instants when the solution was exposed to laser radiation. Activity was measured for a total of 14 days. It is seen that not all the irradiation cycles were accompanied by the same decrease in caesium concentration. This may be due to irreversible changes in the colloidal solution as a result of laser irradiation. Indeed, long-term laser exposure leads to fragmentation of nanoparticles [8–10], thus reducing their size and increasing their concentration. Moreover, it changes laser beam propagation conditions in the solution and laser breakdown plasma parameters.

A result of critical importance is that there is no excess 661-keV gamma radiation during laser irradiation, which is

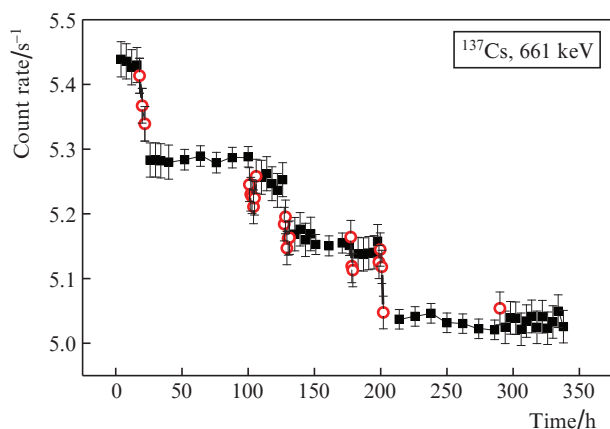


Figure 3. Evolution of the 661-keV activity of a ^{137}Cs solution exposed to periodic 3-ps laser pulses. The filled squares correspond to the time intervals without irradiation and the open circles indicate the instants of laser irradiation.

accompanied by a decrease in nuclide concentration. This suggests that the increased decay rate is due to another channel, differing from conventional spontaneous ^{137}Cs decay with a 30-yr half-life.

Similar experiments were conducted with another laser source: a Nd:YAG laser with a pulse width at half maximum of 10 ns and a pulse repetition rate of 10 kHz. The pulse energy was 2 mJ. The laser beam was focused into a cuvette containing a solution of ^{234}Th and ^{231}Th salts by an F-Theta objective (focal length of 9 cm) and a computer-controlled system of galvanometric mirrors. As a result, the focused laser beam formed a closed loop in the solution, visible by the naked eye, due to the laser breakdown of the solution and breakdown plasma formation along the beam scan path. The typical beam scanning speed was 1 m s^{-1} , which helped to mix the solution. A typical plasma ring is shown in Fig. 4.

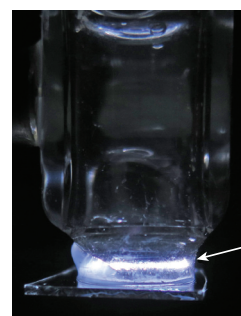


Figure 4. Plasma ring (marked by an arrow) resulting from the optical breakdown of an aqueous solution by a laser beam scanned along a circle. The ring diameter is 8 mm.

The cuvette was placed in the immediate vicinity of the lateral surface of the detector without contact between them. To verify the operation of the detector, an unirradiated sample of another nuclide, whose gamma-ray lines were referenced to the gamma photon energy of the nuclide being irradiated, was placed on its end face.

We studied two beta-active nuclides: ^{234}Th and ^{231}Th . Thorium-234 has a half-life of 28 days and emits 92-keV gamma photons. It forms in an excited state as a result of uranium-238 alpha decay. Uranium of natural isotopic composition contains a small amount (0.7%) of uranium-235, which undergoes alpha decay to thorium-231. This nuclide is also beta-active and emits 186-keV gamma photons. The half-life of thorium-231 is 25.5 h. Both thorium isotopes are situated in the same aqueous solution. The evolution of their concentration during laser irradiation qualitatively resembles that of the caesium-137 concentration (Figs 5, 6). In a number of cases, repeat irradiations produce no changes in activity to within measurement accuracy. Note that the count rate of the unirradiated caesium-134 reference remains constant to within experimental uncertainty, attesting to stable operation of the detector throughout the measurement cycle.

The ^{231}Th activity in the 186-keV line varies in a similar way during sequential laser irradiations. As mentioned above, this peak corresponds to emission from thorium-231, a daughter nucleus of uranium-235. Figure 6 shows the time variation of the count rate for the 186-keV line. As in the case of the laser irradiation of the other beta-active nuclides considered in this study, sequential laser irradiations of the

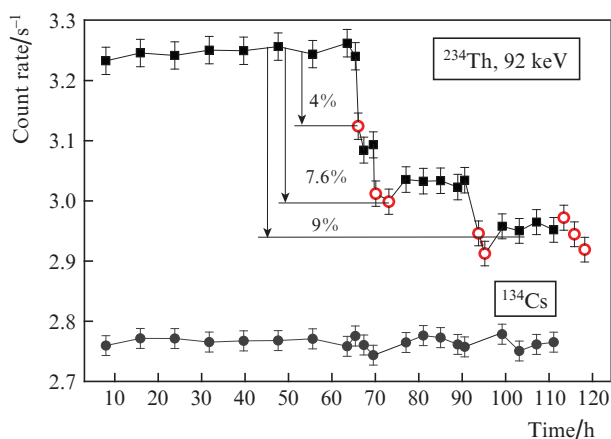


Figure 5. Time variation of 92-keV ^{234}Th activity during repeat exposures of an aqueous uranyl chloride solution to neodymium laser pulses. The open circles indicate the instants of laser irradiation and the filled squares correspond to the time intervals without irradiation. The lower data points (filled circles) represent the activity of an unirradiated ^{134}Cs reference.

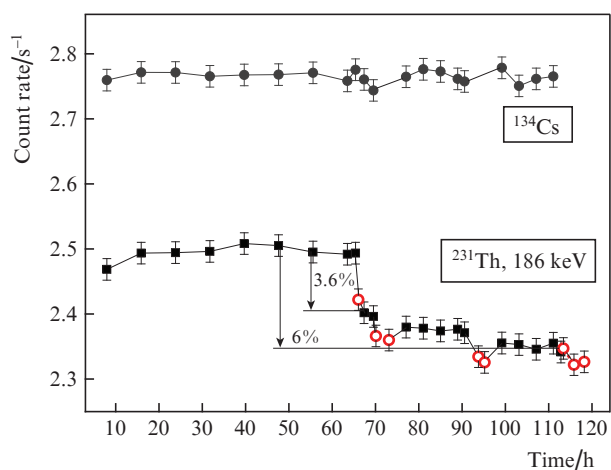


Figure 6. Time variation of 186-keV ^{231}Th activity during repeat exposures of an aqueous uranyl chloride solution to neodymium laser pulses. The open circles indicate the instants of laser irradiation and the filled squares correspond to the time intervals without irradiation. The upper data points (filled circles) represent the activity of an unirradiated ^{134}Cs reference.

solution lead to a nonmonotonic decrease in ^{231}Th concentration, as evaluated using the 186-keV spectral line.

The fact that, in a previous study [7], no increase in ^{231}Th decay rate under laser irradiation was detected is due to the short half-life (25.5 h) of this nuclide, comparable to the time interval between the laser irradiation and activity measurements.

The probability of beta decay to both a bound and a free electron state depends strongly on the electronic structure surrounding the decaying nucleus. For example, Bosch et al. [11] present experimental evidence that complete ionisation of the ^{187}Re nucleus increases the rate of its beta decay to ^{187}Os by many times. The experimental conditions of this study allow us to assume that laser irradiation is accompanied by partial ionisation of hydrated nuclide ions, which may also lead to a considerable decrease in the half-life of the nuclides.

The electromagnetic field of laser radiation is locally enhanced near metallic nanoparticles and nanostructures in liquids [12]. Numerical simulation by Hao and Schatz [13] indicates that the electromagnetic field amplitude enhancement factor near a metallic nanoparticle is 10^2 or greater. In such a field, electrons emitted from the surface of nanoparticles acquire an energy sufficient for the ionisation of surrounding atoms and subsequent plasma formation. If the nanoparticle concentration at the laser pulse waist is sufficiently high, individual plasma regions may overlap, forming a single plasma channel, in which up to 90% of the laser pulse energy will be absorbed. Caesium atoms in the laser pulse waist region can be partially (doubly or triply) ionised by plasma electrons. We also cannot rule out the ionisation of intermediate and lower electron shells in caesium atoms by electrons acquiring sufficient kinetic energy due to the combined action of different mechanisms of electromagnetic field enhancement near nanoparticles and collective effects of electron acceleration in a plasma channel.

Thus, during sequential laser irradiations of aqueous solutions of ^{137}Cs , ^{234}Th and ^{231}Th salts, the activities of all three beta-active nuclides vary in a similar way. The observed decrease in their concentrations under laser irradiation is not accompanied by excess gamma-ray emission in lines characteristic of their spontaneous decay and is due to other mechanisms. Sequential laser exposures of nuclide solutions do not cause a monotonic reduction in the activity of the nuclides, which is due to the evolution of the nanoparticle size distribution and the associated changes in the optical properties of the medium.

Acknowledgements. This work was supported in part by the Russian Foundation for Basic Research (Grant Nos 15-02-04510_a and 16-02-01054_a), the RF President's Grants Council (State Support to Young Scientists of Russia Programme, Grant No. MK-3606.2017.2) and the Presidium of the Russian Academy of Sciences (Grant No. I.25P).

We are grateful to P.A. Chizhov, V.V. Bukin, S.V. Garnov and A.A. Chernov for their assistance in the experimental work.

References

- Andreev A.V., Gordienko V.M., Dykhne A.M., Savel'ev A.B., Tkalya E.V. *Pis'ma Zh. Eksp. Teor. Fiz.*, **66** (5), 312 (1997).
- Andreev A.V., Volkov R.V., Gordienko V.M., Dykhne A.M., Mikheev P.M., Tkalya E.V., Chutko O.V., Shashkov A.A. *Pis'ma Zh. Eksp. Teor. Fiz.*, **69** (5), 343 (1999).
- Schwoerer H., Gibbon P., Düsterer S., Behrens R., Ziener C., Reich C., Sauerbrey R. *Phys. Rev. Lett.*, **86** (11), 2317 (2001).
- Belyaev V.S., Matafonov A.P., Vinogradov V.I., Krainov V.P., Lisitsa V.S., Rousetski A.S., Ignatyev G.N., Andrianov V.P. *Phys. Rev. E*, **72**, 026406 (2005).
- <http://nucldata.nuclear.lu.se/toi/>
- Barmina E.V., Simakin A.V., Shafeev G.A. *Quantum Electron.*, **44** (8), 791 (2014) [*Kvantovaya Elektron.*, **44** (8), 791 (2014)].
- Simakin A.V., Shafeev G.A. *Quantum Electron.*, **41** (7), 614 (2011) [*Kvantovaya Elektron.*, **41** (7), 614 (2011)].
- Mafune F., Kohno J., Takeda Y., et al. *J. Phys. Chem. B*, **105** (22), 5114 (2001).
- Prochazka M., Mojzes P., Stepanek J., Vlckova B., Turpin P.-Y. *Anal. Chem.*, **69** (24), 5103 (1997).
- Kirichenko N.A., Sukhov I.A., Shafeev G.A., Shcherbina M.E. *Quantum Electron.*, **42** (2), 175 (2012) [*Kvantovaya Elektron.*, **42** (2), 175 (2012)].

11. Bosch F., Faestermann T., Friese J., Heine F., Kienle P., Wefers E., Zeitelhack K., Beckert K., Franzke B., Klepper O., Kozhuharov C., Menzel G., Moshhammer R., Nolden F., Reich H., Schlitt B., Steck M., Stöhlker T., Winkler T., Takahashi K. *Phys. Rev. Lett.*, **77** (26), 5190 (1996).
12. Barmina E.V., Lau Truong S., Bozon-Verduraz F., Levi G., Simakin A.V., Shafeev G.A. *Quantum Electron.*, **40** (4), 346 (2010) [*Kvantovaya Elektron.*, **40** (4), 346 (2010)].
13. Hao E., Schatz G.C. *J. Chem. Phys.*, **120** (1), 357 (2004).