

Laser-induced caesium-137 decay

E.V. Barmina, A.V. Simakin, G.A. Shafeev

Abstract. Experimental data are presented on the laser-induced beta decay of caesium-137. We demonstrate that the exposure of a gold target to a copper vapour laser beam (wavelengths of 510.6 and 578.2 nm, pulse duration of 15 ns) for 2 h in an aqueous solution of a caesium-137 salt reduces the caesium-137 activity by 70 %, as assessed from the gamma activity of the daughter nucleus $^{137\text{m}}\text{Ba}$, and discuss potential applications of laser-induced caesium-137 decay in radioactive waste disposal.

Keywords: copper vapour laser, gold nanoparticles, laser ablation, beta decay, radionuclides.

Recent work has shown that the laser irradiation of metal nanoparticles in aqueous solutions of uranium salts causes the activities of the uranium series radionuclides to deviate considerably from their equilibrium values [1, 2]. The activities of the radionuclides vary during the laser irradiation of metallic targets in aqueous solutions of uranium salts, with acceleration of the spontaneous alpha and beta decays of the uranium-238 series nuclides. The effect has been studied in wide ranges of laser wavelengths and pulse durations [3, 4]. The laser ablation of targets in aqueous solutions of salts of nuclides is accompanied by the formation of nanoparticles of the target material in the solution and seems to be a necessary condition for the changes in nuclide activities. This conclusion can be drawn from the fact that the activity of a number of nuclides (thorium-234) rises for a long time, up to 100 days, after laser irradiation [5].

The mechanism underlying the laser-induced increase in the rate of spontaneous decays still remains unclear. There is obvious interest in new experimental data on laser-induced decays of other nuclides, because such data might provide new insights into the fundamental aspects of such processes. In this paper, we present experimental data on laser-induced accelerated caesium-137 decay. In contrast to the uranium-238 series nuclides, caesium-137 has only one decay channel, with a half-life of 30 yr: beta decay to barium-137m. Barium-137m (2.5 min half-life), in turn, decays into barium-137, emitting a gamma photon. Therefore, the gamma emission intensity can be used to assess the caesium-137 con-

tent of a sample and the laser-induced decay rate. An accelerated caesium-137 decay was observed previously, but this nuclide was present as an impurity in a thorium-232 salt [6]. It was therefore of interest to examine the effect of laser irradiation on a solution containing only caesium-137.

In this study, we used a copper vapour laser generating 15-ns pulses at a pulse repetition rate of 15 kHz and wavelengths of 510.6 and 578.2 nm. At an average output power of 4.6 W, the estimated peak power density incident on the target was 10^9 W cm^{-2} . As the target, we used a gold (99.9%) plate. An aqueous solution (2 mL) containing a caesium-137 salt was placed in a cuvette cooled by running water. The target was secured to a specially designed holder and immersed in the solution. The laser beam was focused onto the target from below through a window transparent to it. A typical irradiation time was ~ 1 h. Next, the solution, containing the nanoparticles formed, was poured out of the cuvette. The gamma activity of the solution was measured before and after laser irradiation with an accuracy of $\pm 5\%$ using an Ortec-65195-P semiconductor gamma spectrometer. The activity of the radionuclides present was determined as the area under the corresponding peaks in the gamma spectra of the nuclides. The initial activity corresponded to about 20 μg of caesium-137 in the sample.

The laser irradiation of the target in solution was accompanied by the formation of nanoparticles. The gold particles were elongated, with an average cross-sectional size near 30 nm. As in the case of the laser irradiation of a gold target in a solution containing tritium, a beta-active nuclide [7], the gold nanoparticles in the caesium-137 solution merged to form nanochains, which was accompanied by changes in the absorption spectrum of the colloidal solution. The plasmon resonance of the gold nanoparticles shifted from the green (520–530 nm) to the red (600–650 nm) spectral region, and the enlarged particles rapidly settled to the bottom of the cuvette. The redshift of the plasmon resonance of the gold nanoparticles was caused by a longitudinal plasmon resonance, typical of elongated nanoparticles [8]. As a result, the solution turned blue. Figure 1 is an image of the elongated gold nanoparticles. There are many particles with a large length-to-diameter ratio.

It was of interest to find out when the activity of our samples had changed: directly during the laser irradiation or some time later, as in the case of uranium salts [5]. To this end, the activity was first measured 1.5 h after the laser irradiation. The measurement results are presented in Fig. 2. It is seen that the caesium-137 activity dropped by almost a factor of 4 immediately after the laser irradiation and then remained unchanged for 100 h. This means that the amount of caesium-137 in the sample also dropped by almost a factor of 4

E.V. Barmina, A.V. Simakin, G.A. Shafeev Wave Research Center, A.M. Prokhorov General Physics Institute, Russian Academy of Sciences, ul. Vavilova 38, 119991 Moscow, Russia; e-mail: barminaev@gmail.com, wrc@kapella.gpi.ru, shafeev@kapella.gpi.ru

Received 26 June 2014

Kvantovaya Elektronika 44 (8) 791–792 (2014)

Translated by O.M. Tsarev

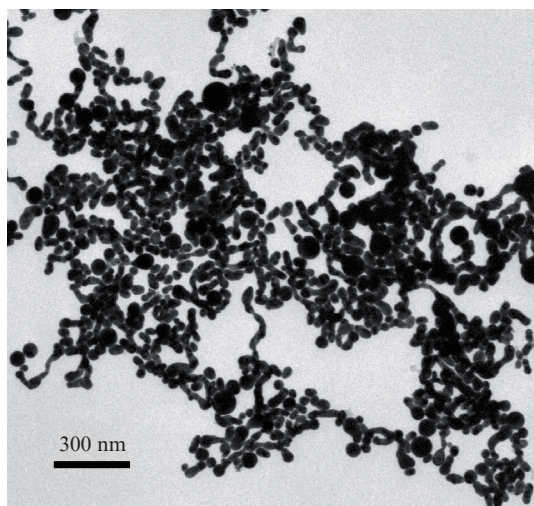


Figure 1. Transmission electron microscopy image of gold nanoparticles produced by the laser ablation of a gold target in an aqueous caesium-137 salt solution.

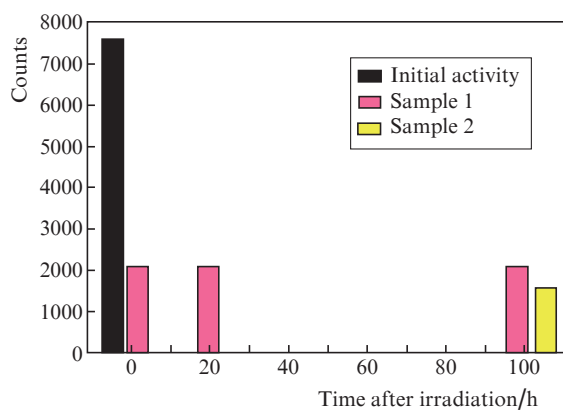


Figure 2. Activity of two samples at 661 keV before and after laser irradiation (counting time, 30 min). The samples were irradiated under identical conditions and had the same initial activity. The laser irradiation time was 1.7 h for both samples.

as a result of the laser irradiation. Additional measurements on various samples 10–20 days after the irradiation showed that the activity of the samples remained unchanged.

The formation of elongated gold nanoparticles in the presence of caesium-137, a beta-active nuclide, seems to be due to the additional charging of the nanoparticles by the electrons emitted by this nuclide. When two spherical nanoparticles accidentally stick together, the charge on them is redistributed along the sticking line and, subsequently, a laser pulse causes them to fuse together. The chain length increases further when such a charged dipole interacts with single spherical nanoparticles. In a sense, this process is the opposite of the laser fragmentation of gold nanoparticles by a copper vapour laser beam [9].

The effect of the electron shell of an atom on the probability that its nucleus will undergo beta decay is well known. For example, absolutely stable in neutral atoms, the ^{163}Dy , ^{193}Ir and ^{205}Tl nuclei are beta-active in fully ionised atoms [10], and full ionisation increases the ^{187}Re beta decay probability by 10^9 times [11]. In the case under consideration, the electron shell of the caesium may experience changes due to the fusion

of the caesium with the gold nanoparticles produced by the laser ablation of the target or the adsorption of caesium ions on the surface of the nanoparticles. In both instances, the electron shell of the caesium can be distorted by plasmon oscillations in the nanoparticles.

Thus, we have presented the first experimental evidence that the exposure of gold targets to a copper vapour laser beam in an aqueous solution of a caesium-137 salt can accelerate the caesium-137 beta decay. The caesium-137 content of the irradiated samples drops immediately after the laser irradiation or during it. The present results extend the list of radionuclides (^{232}Th , ^{238}U , and ^3H) whose decay can be accelerated using laser radiation. The new data are of practical interest: caesium-137 is one of the most dangerous radionuclides because it rapidly spreads through water and soil. The possibility of deactivating it at relatively low laser intensities offers a novel approach to the problem of radioactive waste disposal.

Acknowledgements. This work was supported in part by the RF Ministry of Education and Science through the Support to the Leading Scientific Schools Programme (academician V.F. Bunkin, Grant No. NSH 4484.2014.2).

References

1. Simakin A.V., Shafeev G.A. *Appl. Phys. A*, **101** (1), 199 (2010).
2. Simakin A.V., Shafeev G.A. *Kvantovaya Elektron.*, **41** (7), 614 (2011) [*Quantum Electron.*, **41** (7), 614 (2011)].
3. Barmina E.V., Sukhov I.A., Lepekhin N.M., Priseko Yu.S., Filippov V.G., Simakin A.V., Shafeev G.A. *Kvantovaya Elektron.*, **43** (6), 591 (2013) [*Quantum Electron.*, **43** (6), 591 (2013)].
4. Shafeev G.A., in: *Uranium: Characteristics, Occurrence and Human Exposure*. Ed. by A.Ya. Vasiliev, M. Sidorov (New York: Novapubl. Inc., 2012) pp 117–153.
5. Kirichenko N.A., Simakin A.V., Shafeev G.A. *Phys. Wave Phenom.*, **22** (2), 81 (2014).
6. Simakin A.V., Shafeev G.A. *Phys. Wave Phenom.*, **16** (4), 268 (2008).
7. Barmina E.V., Kuzmin P.G., Timashev S.F., Shafeev G.A. <http://arxiv.org/abs/1306.0830>.
8. Kazakevich P.V., Simakin A.V., Voronov V.V., Shafeev G.A. *Appl. Surf. Sci.*, **252**, 4373 (2006).
9. Kirichenko N.A., Sukhov I.A., Shafeev G.A., Shcherbina M.E. *Kvantovaya Elektron.*, **42** (2), 175 (2012) [*Quantum Electron.*, **42** (2), 175 (2012)].
10. Jung M., Bosch F., Beckert K., et al. *Phys. Rev. Lett.*, **69**, 2164 (1992).
11. Bosch F., Faestermann T., Friese J., et al. *Phys. Rev. Lett.*, **77**, 5190 (1996).