Laser detection of low concentrations of uranium atoms produced in a chemical reaction

V. I. Balykin, V. S. Letokhov, V. I. Mishin, and V. A. Semchishen

Spectroscopy Institute, USSR Academy of Sciences
(Submitted September 21, 1976)
Pis'ma Zh. Eksp. Teor. Fiz. **24**, No. 8, 475–478 (20 October 1976)

The temperature dependence of the concentration of uranium atoms was investigated by the method of laser excitation of fluorescence. The temperature ranged from 1100 to 1800°C.

PACS numbers: 82.50.Et, 82.30.Lp, 42.60.Qm

1. We report here an experimental observation of very small amounts of uranium atoms (10^3 at/cm^3 or ~10 atoms in the registered volume) by exciting fluorescence with a tunable cw dye laser. This method was used to investigate, for the first time, the temperature dependence of the concentration of the uranium atoms produced as a result of the chemical decomposition reactions

$$(NH_4)_2UF_6 \rightarrow 2(NH_4)F + UF_4 \tag{1}$$

and reduction reaction

$$UF_4 + 2Ba \rightarrow U + 2BaF_2 \tag{2}$$

in a high-temperature crucible.

2. The atom-excitation source was a cw dye laser with freely escaping jet, operating with a four-mirror system.[1] The selecting elements were two

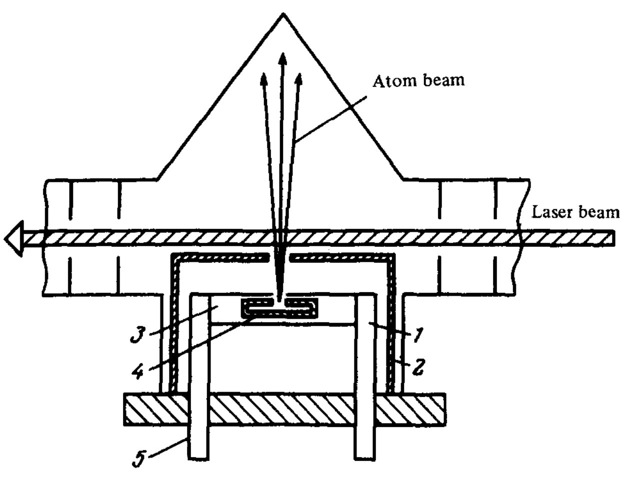

FIG. 1. Cell with vapor source: 1—current leads, 2—heat screen, 3—tantalum foil, 4—crucible.

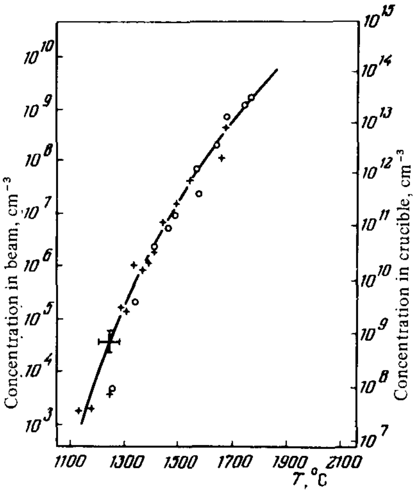

FIG. 2. Dependence of the uranium-vapor concentration on the temperature: left-hand scale—concentration of atoms in the registration zone, right-in the crucible.[2] o-obtained using reactions (1) and (2), +—using metallic uranium.

prisms of heavy flint and a Fabry-Perot etalon in the form of a plane-parallel glass plate. The dye was pumped with an argon laser. The laser wavelength was tuned from 5600 to 6200 Å, the lasing line width was $\approx 2\times 10^{-2}$ Å, and the dye-laser radiation power was $\approx$ 5 mW.

The dye-laser radiation was directed into a cell with uranium-atom vapor (Fig. 1). The source of the vapor was a furnace mounted in the lower part of the cell. The furnace heater was a tantalum foil rolled in the form of a tube. Inside the heating element was placed a tantalum crucible. The crucible was a cylinder with tightly closed cover and with an opening 0.75 mm in diameter on the side surface for the escape of the reaction products. The crucible was heated by passing a current up to 300 A and a voltage 5 V through the foil. The crucible temperature ranged from room to 2000 °C and was monitored with an optical pyrometer. Provision was made in the cell to remove as much as possible the stray light of the pump laser and of the heating element of the furnace. To this end it was blackened, and light screens and diaphragms were placed inside the cell.

We placed in the crucible powdered $(NH_4)UF_6$ and metallic barium. The cell was evacuated to 10^{-4} Torr. When the crucible is heated to 200 °C, the decomposition reaction (1) takes place, and the more volatile compound $(NH_4)F$ evaporates. The uranium reduction reaction (2) was produced at 1100 °C.

The dye-laser emission was tuned to the uranium absorption line $\lambda = 5915.4$ Å. The fluorescence signal was registered by a synchronous detection method. The fluorescence radiation was focused with a lens on the slit of a DMR-4 double monochromator, and an FÉU-79 photomultiplier was placed directly behind the exit slit of the monochromator. The signal from the photomultiplier entered a narrow-band U2-6 amplifier and was registered with a V9-2 synchronous detector.

3. We investigated in the experiment the dependence of the concentration of atoms of the uranium reduced by the chemical reaction, as well as metallic

atoms (in which case metallic uranium was placed in the crucible). The temperature dependence of the uranium-atom concentration is shown in Fig. 2. The left-hand vertical axis indicates the uranium-atom concentration in the registration region, while the right-hand axis shows the concentration of the uranium atoms in the crucible, corresponding to saturated-vapor pressure.[2]

The measurement of the absolute concentration of the atoms was based on a comparison of the fluorescence signals of the uranium atoms and the Rayleigh scattering in air or argon. We used the following numerical values: coefficient of Rayleigh scattering 5×10^{-9} cm^{-1},[3] cross section for the absorption of the laser radiation by uranium atoms 1.1×10^{-13} cm^2,[4] and quantum yield of the fluorescence $\approx 6\times10^{-2}$.[5]

The minimum observable uranium-atom concentration is 3×10^3 at/cm^3, and the minimum detectable number of atoms in the registered volume is about 10 atoms. The threshold for the observation of the atoms was determined in our experiments by the stray radiation from the heating element and the furnace.

The described method of obtaining and registering small concentrations of uranium atoms is suitable for laser spectroscopy of microscopic amounts of transuranic and strongly radioactive elements, as well as for the study of the dependence of the vapor pressure of these elements on the temperature.[6]

In conclusion, the authors are deeply grateful to N. N. Krot for supplying the uranium compound and discussing the results.

[1]W. D. Jonson and P. K. Runge, IEEE J. Quantum Electron. **QE-8**, 724 (1972).
[2]A. N. Zaĭdel', G. V. Ostrovskaya, and Yu. I. Ostrovskiĭ, Tekhnika i praktika spektroskopii (Technique and Practive of Spectroscopy), Nauka, 1976.
[3]I. P. Fabelinskiĭ, Molekulyarnoe rasseyanie sveta (Molecular Scattering of Light), Nauka, 1965. [Plenum, 1968].
[4]Report, Lawrence Livermore Laboratory ERDA UCRL 50021-74, 1974.
[5]Report, Los Alamos Scientific Laboratory LA-4501, Los Alamos, N. M., October 1972.
[6]W. H. Fairbank, T. W. Hänsch, and A. L. Schawlow, J. Opt. Soc. Am. **65**, 199 (1975).