Laser resonant photoionization detection of traces of the radioactive isotope ^{221}Fr in a sample

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The hyperfine splitting of the D_2 line of the isotope ^{221}Fr ($T_{1/2} = 4.8$ min) has been measured. The ionization potential of the francium atom has been refined: $E_i \leqslant 4.154$ eV. The experiments were carried out at a francium flux of 8×10^2 isotopes/s from the sample. The sensitivity of the detection of Fr in a sample is 10^4 atoms.

1. Laser resonant photoionization of ions is one of the most sensitive methods for detecting particles.[1] The high sensitivity of this method makes it possible to study the hyperfine structure and isotopic shifts of atomic lines at atomic fluxes of 10^4–10^5 s^{-1}, corresponding to an average of about 1–10 atoms in the observation region.[2] There is

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considerable interest in the detection in samples of, and the study of, atoms of rare radioactive elements.[1)] One such element is francium (Fr). The decay half-life of the longest-lived isotope, ^{223}Fr, is 22 min, so that in natural samples it would be present in extremely small quantities, as the product of the radioactive decay of U atoms. As a result, the first spectral studies of Fr became possible only in the late 1970s, with the help of an accelerator, at fluxes[4] $\sim 10^8$ atoms/s. At present, the wavelengths of transitions from the ground state to four low-lying states have been measured.[5]

In the present paper we report the results of spectral studies of, and the detection of, Fr atoms at fluxes 10^5 times smaller than those in Ref. 4. This advance opens up some fundamentally new opportunities in the atomic spectroscopy of radioactive elements, making it possible to work with an activity of less than 10^{-6} Ci under standard laboratory conditions.

2. The test sample contained $\sim 10^9$ ^{225}Ra atoms, implanted at a depth ~ 100 Å in a tantalum foil 10×20 mm in area and 10 μm thick. This sample was synthesized by collecting the ^{225}Ra recoil nuclei formed as a result of the radioactive decay of ^{229}Th deposited as a thin film on the tantalum foil. The radioactive decay of the ^{225}Ra atoms generates a chain of elements,

$$\xrightarrow[14.8\text{ days}]{\beta} {}^{225}\mathrm{Ac} \xrightarrow[10\text{ days}]{\alpha} {}^{221}\mathrm{Fr} \xrightarrow[4.8\text{ min}]{\alpha},$$

one of which is ^{221}Fr. The steady-state amount of ^{221}Fr in the sample is $N_{ss} = nT_{1/2}/\ln 2 = 3.3\times 10^5$ atoms, where $n = 8\times 10^2$ s^{-1} is the rate at which ^{221}Fr is generated as a result of the radioactive decay of ^{225}Ac (the measurements were based on the α activity).

Figure 1 shows the arrangement used to photoionize the Fr. The beam from a copper-vapor laser[6] 1 (the energy in the pulse is 0.65 mJ, the pulse length is $\simeq 15$ ns, and the pulse repetition frequency is 8.7×10^3 Hz) is split in two by a dielectric mirror. One of the resulting beams, with a wavelength of 578.2 nm (40%), is used to pump dye laser 2, whose output ($\lambda_1 = 718.0$ nm) performs the excitation in the first step; the second beam, with a wavelength of 510.6 nm, is used to ionize the atoms from the excited state. The pulses of the two beams are synchronized with the help of an optical

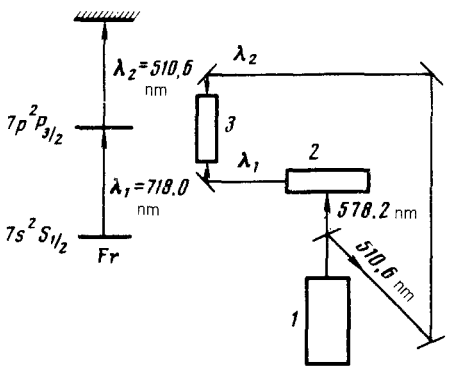

FIG. 1. Schematic representation of the laser photoionization of Fr. 1—copper-vapor laser; 2—dye laser; 3—ionization chamber.

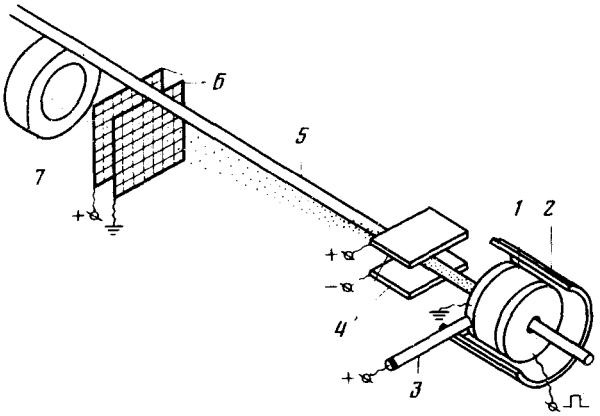

FIG. 2. Layout of the ionization apparatus and the detection system. 1—ionization cavity; 2—heater; 3—evaporator; 4—deflecting plates; 5—laser beams; 6—electrostatic analyzer; 7—secondary electron multiplier.

delay of the light at $\lambda_2 = 510.6$ nm. The laser beams meet, moving in opposite directions, in ionization chamber 3.

Figure 2 shows the layout of the ionization apparatus and the detection system. The Fr atoms are photoionized in heated cavity 1 (~ 600 °C), formed by two titanium expelling plates, 30 mm in diameter, separated by an insulator—a quartz tube 10 mm long with a wall thickness of 2 mm. The cavity is heated by the radiation from hot surface 2. The Fr atoms are evaporated from the sample in tantalum branch pipe 3 at a temperature $T \simeq 1250$ °C. They enter the ionization cavity through a channel in one of the expelling plates. Metallic Ba is held in the cavity to passivate the walls of the cavity and to reduce the work function of the materials from which the cavity is made. As the atoms undergo random walks in the cavity, they repeatedly cross the ionization region, with the result that the probability for their passage through the laser beam and thus their ionization efficiency are increased.[7] The resulting ions are expelled from the cavity through an aperature 1 mm in diameter by an electric field pulse with a height of 1.1 kV/cm and a length of 350 ns. These ions are then deflected by plates 4 and detected by a VÉU-7 multichannel electron multiplier 7. In order to reduce the background resulting from thermal ions, we carry out a mass and energy analysis of the ions, making use of the time of flight (the drift region is 50 cm long) and an electrostatic analyzer in the form of grids (6). The vacuum in the chamber is 10^{-7} torr.

3. The Fr atoms are identified on the basis of the resonant peak in the photocurrent at the wavelength 718.0 nm, which arises during the tuning of the frequency of the laser of the first step. The time-of-flight mass analyzer is tuned to the mass corresponding to Fr atoms (Fig. 3) for these measurements. The hyperfine structure of the D_2 line ($\lambda = 718.0$ nm) is studied with a laser with an output spectral width of 0.3 cm^{-1}. The splitting of the hyperfine components found experimentally is 0.61 ± 0.02 cm^{-1}, in agreement with the value of 0.62 cm^{-1} found in Ref. 5, but for a different line (D_2', $\lambda = 422.57$ nm). This agreement of hyperfine splittings is further proof of the detection of francium.

The total energy of the laser photons which ionize the francium atoms in the resonant fashion is 4.154 eV. It follows that the ionization potential of francium is

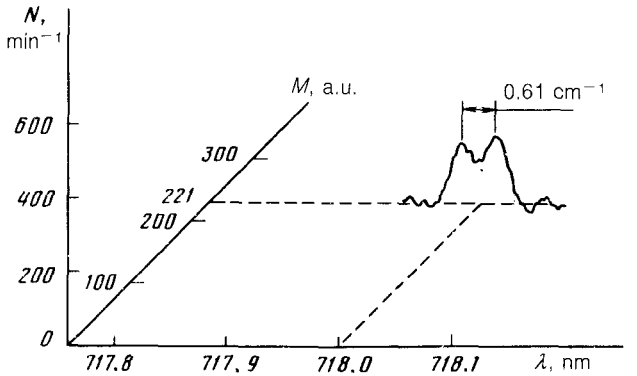

FIG. 3. Identification of the Fr atoms. This spectrum was obtained at a flux of 8×10^2 atoms/s.

$E_i \leqslant 4.154$ eV (the probability for the frequency of the copper laser to fall at a transition to a Rydberg state is small). The value which we find for the ionization potential is lower than that given in Ref. 4 ($E_i = 4.19$ eV).

4. The sensitivity of the detection of the Fr atoms in a sample was determined by the following procedure. For a time $4T_{1/2}$, the francium was accumulated in a cold branch pipe. This pipe was then rapidly heated. The heating conditions were chosen to evaporate the Fr in 1 min. Over this time, we detected 750 counts after subtraction of the background. This number of counts corresponds to a certain steady-state amount of ^{221}Fr in the sample: 3.3×10^5. It follows that the efficiency at which the Fr atoms are detected is 2×10^{-3}. It is a simple matter to determine the detection sensitivity, by noting that the background fluctuations in this experiment amounted to $\simeq$35 min^{-1}. At a unit signal-to-noise ratio, the sensitivity would be 1.5×10^4 Fr atoms in the sample.

5. This study shows that there is an alternative approach to the spectroscopy of elements which nature has not provided us in sufficient amounts for optical studies. Instead of using expensive accelerator installations to generate large fluxes of these elements,[4,5] one can make use of slightly radioactive sources of these elements, combined with ultrasensitive photoionization laser spectroscopy, which is accessible today to any laboratory.

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[1)]The idea of resonant photoionization detection of individual atoms of radioactive elements was advanced in Ref. 3.

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