Laser optical spectroscopy on francium D_2 resonance line

S. Liberman, J. Pinard, H. T. Duong, P. Juncar, P. Pillet, J.-L. Vialle, and P. Jacquinot
Laboratoire Aimé Cotton, Centre National de la Recherche Scientifique II, 91405 Orsay, France

F. Touchard, S. Büttgenbach,* C. Thibault, M. de Saint-Simon, and R. Klapisch
Laboratoire René Bernas du Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse, 91406 Orsay, France

A. Pesnelle†
CERN, Geneva, Switzerland

G. Huber
Gesellschaft für Schwerionenforschung, D 6100 Darmstadt 1, Germany
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A highly sensitive method of detection coupled with a laser atomic beam experiment using on-line-produced Fr isotopes, has permitted finding and measuring the first optical resonance line of this element and its wavelength: $\lambda = 717.97 \pm 0.01$ nm. A high-resolution optical study has been undertaken, which has led to the determination of the hyperfine structure and isotope shifts for isotopes of mass number 208 to 213.

I. INTRODUCTION

Among the heaviest elements which can be found in nature, francium was, until recently,[1] the only one for which no optical transition had ever been observed. Since its discovery in 1939,[2] many experiments have failed in observing these optical transitions. One may notice first that the longest-lived isotope ^{223}Fr, which corresponds to the natural one, has a half-life of 22 minutes and occurs in a weak (1.4%) α-decay branch of ^{227}Ac. Natural radioactivity thus gives rise to small quantities of francium atoms, corresponding at equilibrium to a total amount of less than 30 g of that element present at any moment in the earth's crust.

In addition the classical methods of optical spectroscopy have proved to be somewhat inadequate for this problem; on the one hand, in a cell, francium atoms would be associated with several other much more abundant radioactive species which would lead, in emission spectroscopy, to such an intricate spectrum that it would be hopeless to attribute one line to the francium element. On the other hand, chemical agressivity of this element would cause it to be adsorbed on the walls of the cell; its density would rapidly diminish in the active volume, and this would prevent success in a classical optical absorption experiment.

A new possibility arose recently at the CERN-ISOLDE on-line mass separator, which makes available a source of about 10^8 atoms per second of chemically and isotopically pure Fr isotopes,[3] by means of the spallation of uranium by 600-MeV protons. Such an amount of 10^8 atoms is quite sufficient for an experiment using laser atomic beam spectroscopy in a way similar to the one reported for the hyperfine spectroscopy of the long series of alkali atoms.[4] Let us recall that basically, the experiment uses singly ionized atoms delivered by the ISOLDE facility,[3] which are converted into thermal neutral atoms of an atomic beam after implantation on, and reevaporation from, the surface of an yttrium-coated tantalum tube heated to 1200 °C. Then the atoms of the beam interact through their resonance line with the light of a laser beam propagating perpendicularly to the direction of the atomic beam. For high-resolution purposes as was the case for Na (Ref. 4), Cs, (Refs. 4 and 5), and Rb (Ref. 5) isotopes, the laser light is provided by a single-mode tunable cw dye laser. The interaction with light induces an optical pumping between the two hyperfine sublevels of the ground state, and changes the relative populations of the magnetic sublevels corresponding to $m_J = +\frac{1}{2}$ and $-\frac{1}{2}$. These modifications are analyzed by a six-pole magnet which focuses atoms with $m_J = +\frac{1}{2}$ and defocuses the others, provided the atoms experience a strong-field region inside the magnet. The detection system is formed with a mass spectrometer and an ion counter.

The first step in such a program of Fr optical spectroscopy is obviously to locate the resonance line. Figure 1 summarizes the different predictions for the value of the wavelengths of the first two resonance lines, which are derived either from empirical extrapolations or from *ab initio* calculations. It appears that a reasonable range to be explored in order to find the first resonance transition (D_2 line) would extend be-

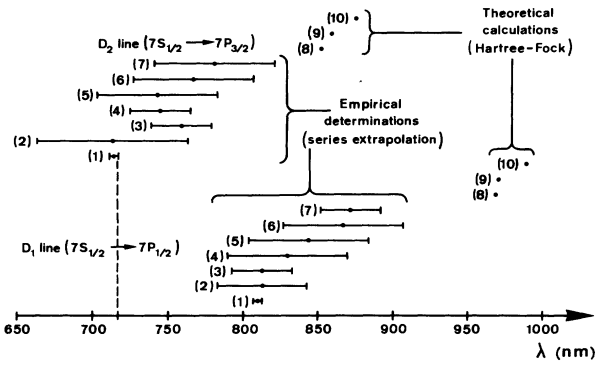

FIG. 1. Theoretical and semiempirical predictions of the wavelengths of D_1 and D_2 Fr lines with their estimated uncertainty bars. Points (1) and (2) correspond to extrapolation of the last elements of the alkali series compared to the Cu, Ag, Au series. This extrapolation is rather well established by comparison of correspondent singly ionized elements (Sr^+, Ba^+, Ra^+, and Zn^+, Cd^+, Hg^+). The other points are obtained by extrapolation of quantum defects all along the alkali series and singly ionized alkali-earths. (1) See Ref. 7; (2) J. F. Wyart (private communication); (3) K. T. Bainbridge, Phys. Rev. 34, 752 (1929); (4) H. Yagoda, Phys. Rev. 38, 2298 (1931); (5) the authors of present paper; (6) and (7) A. C. Muller, diplomarbeit, Mainz, 1977 (unpublished); H. J. Kluge (private communication); (8) and (9) M. Aymar (private communications); (10) J. P. Desclaux, thèse, Paris, 1971 (unpublished).

tween 700 and 800 nm. A suitable dye (oxazine 725) can be found for that spectral region. In order to find the line in a period of time compatible with reasonable experimental running conditions, it was thus necessary in a first experiment to use a low-resolution laser system. This experiment, which has permitted finding the Fr D_2 line and measuring its wavelength, is described in Sec. II. Having then located the position of that first resonance line, it became possible to perform high-resolution observations, enabling us to measure hyperfine structures and isotope shifts for several isotopes. This high-resolution part of the study is described in Sec. III.

II. SEARCH FOR AN OPTICAL TRANSITION IN *Fr*

As stated previously, in a low-resolution phase of the experiment a broadband excitation system is needed associated with a magnetic detection of optical resonances. It has been achieved using our already described experimental arrangement,[4] by introducing two major modifications. The first one is an angular spread of the light beam along the atomic beam, produced by means of cylindrical lenses (see Fig. 2). In so doing the absorption profile of the atoms is Doppler broadened and all

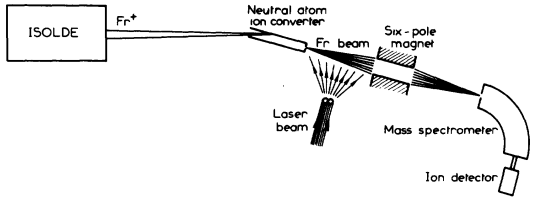

FIG. 2. General scheme of the experimental arrangement. Fr^+ ions are delivered by the ISOLDE facility with an energy corresponding to 60 keV. They are neutralized and thermalized by impinging on a hot tantalum tube coated with a thin layer of yttrium.

of them are excited, whatever their longitudinal velocity. The second modification, and the more important one, concerned the laser system itself. The laser configuration which we used is shown in Fig. 3. The laser cavity is of the symmetric type, which facilitates multimode oscillation. Since the cavity length is about 50 cm, longitudinal modes are spaced at about 300 MHz. The laser cavity is closed at one end by a totally reflective mirror and at the other by a highly efficient reflection grating. Close to the grating a glass plate, antireflection coated on one face and partially reflective on the other face, is placed perpendicularly to the laser axis in such a way that the system composed by the grating and the glass plate plays the role of a Fabry-Perot interferometer. The thickness of the equivalent interferometer corresponds to specific modes spaced by approximately $\Delta\sigma = 9$ GHz. The whole cavity is adjusted to oscillate in several modes, giving a laser frequency pattern [Fig. 3 (b)] formed of five or six packets of a few consecutive modes, each packet being spaced from the following one by $\Delta\sigma$. Continuous scanning operation is achieved by a sawtooth voltage applied

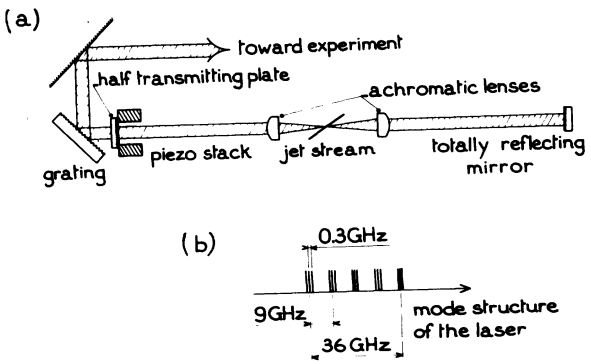

FIG. 3. (a) General scheme of the laser cavity arrangement; (b) mode structure of the laser. Displacing such a mode spectrum by 9 GHz results in scanning a range of 45 GHz.

to the glass-plate piezoceramic, which linearly shifts the laser mode pattern by about $\Delta\sigma$; it actually corresponds to a scanned frequency range of five to six times $\Delta\sigma$ and consequently reduces the recording time by the same factor. As the piezoceramic voltage decreases to zero, the grating is rotated (using a step-by-step motor) in order to jump a frequency interval of 45 GHz. Concerning the magnetic detection technique of the optical resonances which is used in these experiments, one has to pay attention to the fact that the efficiency of this method strongly depends on whether or not the atoms experience a "strong" magnetic field. The magnetic field is said to be strong as soon as it decouples I and J, or in other words when the Zeeman energy splitting in the ground level is larger than the hyperfine splitting. Since the maximum value of the field at the pole pieces of the six-pole magnet is 9 kG, one calculates that the hyperfine splitting of the ground state should not exceed 25 GHz. In fact, as will be seen later on, the ground-state hyperfine splitting greatly exceeds this value; however, using Zeeman optical pumping with such an intermediate field strength by means of properly polarized exciting light, it is still possible to detect all the hyperfine components of the line structure.

The flux of francium ions finally reaching the electron multiplier is about 10^3/s off resonance, i.e., a transmission rate of 10^{-5} for the whole system. Earlier experiments had shown that radioactive atoms implanted in the first dynode of an electron multiplier produced spurious pulses due to recoils from radioactive decay. The accumulation of francium and its descendants would then quickly contaminate the electron multiplier. For that reason, a special detector in which the first dynode was made of aluminized Mylar tape was built.[6] The secondary electrons are focused on a conventional Cu-Be electron multiplier while the tape can be moved at will, freeing the surface of any radioactivity. After a predetermined counting time the tape was moved by a step motor. In this way the background at the multiplier was not more than 10%.

During the laser scanning, the light beam is mechanically chopped at a frequency of 2 Hz. The two corresponding ion signals (with and without exciting light) are stored in two buffer counters in which they are accumulated during preset periods of time which are adjusted between 1 and 10 s, depending upon the production rates of Fr atoms (the mean number of counted ions must be around 2000). This procedure gives rise to an intensity-normalized spectrum, which permits elimination of the fluctuations in Fr atoms production due to the variation of the intensity of the proton beam

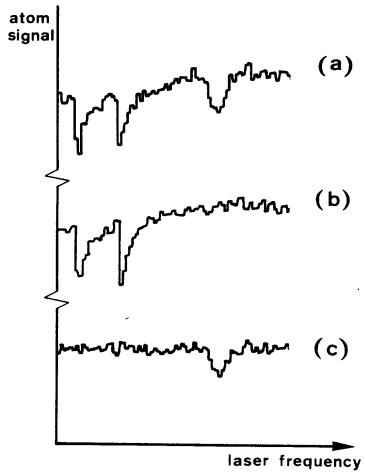

FIG. 4. Typical recordings of the ion signal versus laser frequency. Trace (a) gives the signal with laser light; trace (b) gives the signal without laser light; trace (c) gives the normalized signal obtained by subtracting signal (b) from signal (a). The remaining resonances are due to the Fr atoms.

which hits the uranium target. A typical recording is given in Fig. 4. It shows the resonance signal which was observed after starting from 700 nm and scanning the laser frequency toward increasing wavelengths. This allowed photographing of the laser light spectrum tuned at the resonance together with a neon reference spectrum through a grating spectrograph. The result is shown in Fig. 5. It permits one to distinguish the two components corresponding to the hyperfine splitting of the ground state and therefore to measure both the wavelength of the Fr D_2 line and the order of magnitude of the above-mentioned hyperfine splitting. It has been found $\lambda(D_2) = 717.97 \pm 0.01$ nm, and $\Delta(\text{hfs}) \simeq 50$ GHz. It is quite remarkable that this wavelength value is in good agreement with the empirical determination done by H. Yagoda (see Fig. 1) in 1932,[7] even before the discovery of the Fr element. Notice that it matches several other determinations and in particular a recent *ab initio* calculated value.[8]

FIG. 5. Photograph of the laser light spectrum when it is tuned on the Fr D_2 resonance line. It is to be emphasized that in order to increase the precision of the wavelength measurement this picture has been taken using a dye laser oscillating on one single mode. This procedure has permitted us to distinguish between the two components due to the hyperfine splitting of the ground state.

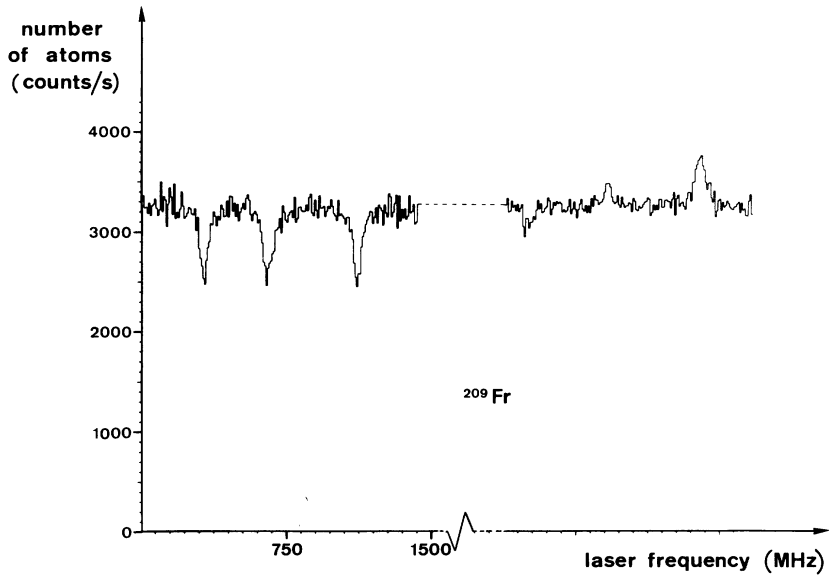

FIG. 6. Hyperfine-structure recording of ^{209}Fr D_2 resonance line. The two groups of three components are about 50 GHz apart. The presence of a negative component in the group on the right-hand side is explained in the text.

III. HIGH-RESOLUTION HYPERFINE-STRUCTURE STUDY

Having located the frequency of the Fr D_2 line, a high-resolution study was undertaken. It used the same experimental set-up as the one used for the other alkali atoms[4,5] and in particular the same laser arrangement of a CR-599 dye laser, pumped by a Kr-ion laser frequency driven by a sigmameter.

A typical recording of the hyperfine structure of the D_2 line of the Fr isotope of mass number 209 is given in Fig. 6. As previously mentioned in Sec. II, this recording is different from the ones obtained for the other alkalis; in particular, one can observe the transition $(F = I - \frac{1}{2}) \rightarrow (F' = I - \frac{3}{2})$ in the D_2 line. This type of signal can be detected only in the low magnetic-field case using Zeeman optical pumping with σ-polarized light, because the magnetic sublevels of the $F = I - \frac{1}{2}$ hyperfine level have different magnetic moments. The effective magnetic moment of the initial and final state of the atom can differ by approximately one Bohr magneton, allowing us to detect this ordinarily unobservable transition. Due to the huge hyperfine splitting of the ground state, it was not possible to record the complete structure within one single scanning range of the laser (this range is actually limited to about 35 GHz continuous scanning). Therefore, in order to measure the whole structure, it was necessary to stop the frequency scanning and then to manually shift the laser frequency to a known amount corresponding to the free spectral range $\delta\sigma$ of the intracavity thick etalon, before pursuing the remaining frequency exploration. This $\delta\sigma$ value has been experimentally evaluated thanks to the sigmameter and found to be $\delta\sigma = 10.00$ GHz with a reproducibility of about 50 MHz. Of course, this procedure lowers somewhat the overall accuracy of the measurements in comparison with the other long series of alkali isotopes that we studied by the same methods. On the other hand it has not only permitted us to measure the hyperfine structure of the six Fr isotopes, but also to locate the center of gravity of each isotope with respect to the others. A schematic display of all the recorded structures is reported in Fig. 7. Table I gives the corresponding values of the isotope shifts (using neutron magic ^{213}Fr as a reference), as well as the hyperfine constants of the ground state ($^2S_{1/2}$) and of the excited state ($^2P_{3/2}$).

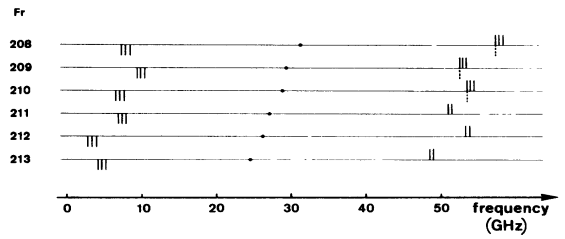

FIG. 7. Schematic display on a common frequency scale of the recorded structures of isotopes of mass number 208 to 213. The dot indicates the center of gravity of each structure.

TABLE I. Nuclear and atomic characteristics of Fr isotopes of mass number $A(=208-213)$. Nuclear spins I have been taken from Ref. 11. Isotope shifts (IS) are measured referring to isotope 213. Magnetic hyperfine constants A and electric quadrupole constants B are given for both ground level $7s\,^2S_{1/2}$ and excited level $7p\,^2P_{3/2}$. Quoted errors correspond to one standard deviation.

A	I	IS (MHz)	$A(^2S_{1/2})$ (MHz)	$A(^2P_{3/2})$ (MHz)	$B(^2P_{3/2})$ (MHz)
208	7	6645.2 (9.2)	6639.7 (7.0)	72.8 (0.5)	8.9 (7.5)
209	$\frac{9}{2}$	4771.4 (7.5)	8590.5 (10.5)	93.1 (0.6)	−61.0 (5.8)
210	6	4243.1 (8.0)	7182.4 (8.1)	77.9 (0.2)	47.6 (2.2)
211	$\frac{9}{2}$	2533.7 (8.0)	8698.2 (10.5)	94.7 (0.2)	−55.3 (3.4)
212	5	1628.3 (7.5)	9051.3 (9.5)	99.1 (0.9)	−35.3 (15.5)
213	$\frac{9}{2}$	0	8744.9 (10.5)	94.5 (1.6)	−20.7 (17.0)

It is to be noted that the value of the ratio $A(^2S_{1/2})/A(^2P_{3/2})$ is constant to better than 0.5%, which is an indication of the consistency of the measurements. No hyperfine anomaly has been found for the studied isotopes. From the above experimental values it is not possible to extract any nuclear moment. An ABMR experiment is in progress to measure g_I (Ref. 9). In fact, there exists a series of g-factor measurements of the $(h_{9/2})^n$ proton states of $N=126$ nuclides. In the case of ^{213}Fr, for instance, one finds $g_I=0.888(3)$ for the state $(h_{9/2})^5_{I=21/2}$.[10] Assuming that g_I is independent of I and n, it is possible to calculate a plausible value of the magnetic moment $\mu(^{213}\mathrm{Fr})=3.996(14)\mu_N$. As a check, one should note that $^{209}_{83}\mathrm{Bi}_{126}$ and $^{213}_{87}\mathrm{Fr}_{126}$ have the same neutron number and their unpaired proton is in the same $h_{9/2}$ level, since both have a nuclear spin $\frac{9}{2}$. Therefore their magnetic moments are expected to be very close. The measured value of $\mu(^{209}\mathrm{Bi})$ is 4.1 μ_N,[12] which is in excellent agreement with the previously calculated value of $\mu(^{213}\mathrm{Fr})$. Using this value it is easy to deduce from the measured value $A(^2P_{3/2})$ of ^{213}Fr and the usual Fermi-Segre formula a new evaluation of the $7p$ fine-structure splitting, $\Delta T=1573$ cm^{-1}. This leads to a semi empirical determination of the wavelength location of the D_1 line which, under these assumptions, would lie at $\lambda(D_1)=809.4 \pm 10$ nm. This value is to be compared to the ones given in Fig. 1. The effective quantum number of the ground state, $n^*(^2S_{1/2})$, can also be calculated using the measured hyperfine coupling constant $A(^2S_{1/2})$ of ^{213}Fr and the Fermi-Segre formula. We obtained $n^*=1.802$ placing the ionization limit at 33 800 cm^{-1} (4.19 eV).

IV. CONCLUSION

This first series of experiments on Fr isotopes has permitted us to measure the wavelength of the D_2 line and to study the hyperfine structure and isotope shift of six isotopes. Obviously, new experiments are needed in order to find out the location of the D_1 line and even to locate other resonance lines, such as $7s-8p$ lines, which are expected to lie in the blue range of the visible spectrum. Concerning high-resolution investigations the study of heavier isotopes of Fr would be of great physical interest because it would correspond to the first systematic isotope measurements across the 126 neutron magic number.

*Present address: University of Bonn, Germany.
†Service de Physique Atomique, Centre d'Etudes Nucléaires, Saclay, France, and the "Isolde" collaboration.

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