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H.T. Duong, S. Liberman, J. Pinard, A. Coc, C. Thibault, et al.. Accurate determination of ground state hyperfine structures of some radioactive alkali isotopes by r. f. magnetic resonance and laser optical pumping. *Journal de Physique*, 1986, 47 (11), pp.1903-1908. 10.1051/jphys:0198600470110190300 . jpa-00210386

HAL Id: jpa-00210386

<https://hal.science/jpa-00210386>

Submitted on 4 Feb 2008

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Classification
Physics Abstracts
32.30-35.10

Accurate determination of ground state hyperfine structures of some radioactive alkali isotopes by r.f. magnetic resonance and laser optical pumping

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(Reçu le 27 mai 1986, accepté le 21 juillet 1986)

Résumé. — Les structures hyperfines de l'état fondamental de $^{76,77,82,84m}\text{Rb}$, ^{140}Cs et ^{226}Fr ont été mesurées avec précision par une méthode associant la résonance magnétique et le pompage optique laser. Par rapport aux techniques purement optiques la précision des mesures est améliorée par trois ordres de grandeur. Les spectres obtenus pour l'isotope le moins abondant (^{76}Rb) donnent une bonne idée de la sensibilité du montage. La comparaison en terme de sensibilité, avec le dispositif classique de Rabi est en faveur de la méthode présente.

Abstract. — The ground state hyperfine structures of $^{76,77,82,84m}\text{Rb}$, ^{140}Cs and ^{226}Fr have been accurately measured by an atomic beam magnetic resonance method associated with laser optical pumping. In comparison with the purely optical methods a gain in accuracy of three orders of magnitude is achieved. Spectra obtained for the least abundant isotope (^{76}Rb) give a good estimate of the sensitivity of the set up, which is favourably compared to the one obtained with classical ABMR Apparatus.

1. Introduction.

Since the first experiment in 1975 [1] high resolution laser spectroscopy on thermal beams associated with a very sensitive magnetic detection has proved to be a fruitful method for the investigation of nuclear properties on long series of radioactive alkali isotopes. Isotope shifts (I.S.) and hyperfine constants ($A(^2\text{S}_{1/2})$, $A(^2\text{P}_{1/2})$, $A(^2\text{P}_{3/2})$, $B(^2\text{P}_{3/2})$) have been measured by optical spectroscopy with an accuracy of a few MHz, even 1 MHz in the best cases, for Na, K, Rb, Cs and Fr [2-6] isotopic sequences.

Concerning the hyperfine constants, it is well known that radiofrequency techniques may lead to

more accurate determinations than optical spectroscopy. For the excited state hyperfine constants, the natural lifetime limits the gain in accuracy but for the hyperfine structure of the ground state the frequency width of the resonances is only governed by the interaction time. Precisions better than 1 Hz have been reported [7, 8] and an accuracy of a few kHz is easily achievable. Therefore, we can get a gain in accuracy by three orders of magnitude for the determination of the $A(^2\text{S}_{1/2})$ hyperfine constants by performing an additional r.f. magnetic resonance experiment. This has been demonstrated on the ^{43}K and ^{44}K isotopes [9]. We report here on a similar experiment performed on Rb, Cs and Fr isotopes in parallel with laser optical spectroscopy experiments [10], [6]. Due to the frequency limitation of the available r.f. excitation (< 2 GHz in our case) only six isotopes have been studied. The least abundant one (^{76}Rb) provides a good test of the sensitivity of the method.

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2. Principle of the experiment.

The principle of the laser r.f. spectroscopy experiment has been extensively explained in a previous paper [9] and we shall recall it only briefly.

A thermal atomic beam of the studied isotope interacts at a right angle with the light beam delivered by a c.w. tunable single mode dye laser in the presence of a weak static magnetic field H_0 (see Fig. 1). At resonance, optical hyperfine pumping modifies the relative populations of the ground state magnetic substates (F, m_F) . These population changes are analysed by a sixpole magnet which focusses the atoms with $m_j = +1/2$ and defocusses those with $m_j = -1/2$. The focussed atoms are ionized, mass separated and counted (see Figs. 1 and 2 in Ref. [4]). When the laser frequency is scanned, the optical resonances appear as negative or positive peaks on a mean ion beam intensity according to whether the optical pumping process increases or decreases the number of focussed atoms. One can get, this way, a first determination of the hyperfine ground state splitting with an accuracy of a few MHz owing to the residual Doppler width of the observed optical transitions (typically 30 MHz) and to the precision and the reliability of the laser frequency control ensured by the sigmameter [11].

The laser frequency is then locked on an atomic transition corresponding to a negative peak and the laser light is σ^- polarized with respect to H_0 in order to maximize the amplitude of this peak. Downstream from the laser interaction region and before they enter the sixpole magnet, the optically pumped atoms interact with a r.f. magnetic field. When its frequency is resonant with an allowed magnetic transition between two Zeeman sublevels, the interaction tends to equalize the population of the two involved sublevels and the detected ion signal increases again. It is then possible to observe the following high-frequency magnetic transitions

$$\left(F = I - 1/2, m_F \leftrightarrow F = I + \frac{1}{2}, m_F + \Delta m_F \right)$$

with $\Delta m_F = \pm 1$ if H_0 and the r.f. field are perpendicular, $\Delta m_F = 0$ if they are parallel. Finally the hyperfine splitting in zero field

$$(F = I - 1/2 \leftrightarrow F = I + 1/2)$$

is easily deduced from the measured frequencies of the observed transitions.

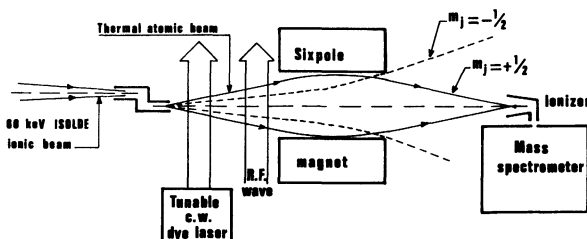


Fig. 1. — Experimental set-up.

Table I. — Measured ground state hyperfine constants.

Isotope (I)	$\Delta \nu_{\text{HFS}}$ (MHz)	$A(^2S_{1/2})$ (MHz)	
		This work	Other results
^{76}Rb (1)	1 041.1674 (40)	694.1116 (27)	701 (17) ^(a)
^{77}Rb (3/2)	1 625.5004 (40)	812.7502 (20)	815.7 (51) ^(a) 812.5 (35) ^(b)
^{82}Rb (1)	1 549.3847 (30)	1 032.9231 (20)	1 031 (10) ^(a) 1 031.6 (11) ^(c)
$^{84\text{m}}\text{Rb}$ (6)	429.6999 (20)	66.10768 (31)	68.2 (46) ^(a)
^{140}Cs (1)	625.6684 (20)	417.1123 (14)	417.6 (22) ^(d) 417.1 (39) ^(e) 417.6 (5) ^(f)
^{226}Fr (1)	1 047.1606 (30)	698.1071 (20)	699 (4) ^(g)

^(a) Ref. [4]

^(b) Ref. [16]

^(c) Ref. [19]

^(d) Ref. [5]

^(e) Ref. [17]

^(f) Ref. [18]

^(g) Ref. [6].

3. The experimental set-up.

The apparatus is approximately the same as the one used for optical laser spectroscopy and we will go into more details only for the particular modifications introduced for this r.f. spectroscopy experiment. The studied radioactive isotopes are produced and separated in mass by the ISOLDE on line facility at CERN.

A detailed description of the atomic beam apparatus and of the data acquisition system is given in references [4] and [12]. Informations concerning the production yields of the radioactive isotopes and the dye laser system to be used for each element can be found in references [4-6] for respectively the rubidium, caesium and francium isotopes.

The interaction region has been slightly modified since the first r.f. experiment on potassium isotopes [9]. It is shown schematically in figure 2a. The r.f. electromagnetic wave propagates in a 50 Ω coaxial line, the section of which changes progressively from a circular shape (usual r.f. cable) to a rectangular one and again to a circular one (see Fig. 2b). The atomic beam intersects this r.f. line at a right angle in its central part where the external conductor presents a rectangular section and the internal one is a flat metallic plate. The sizes of these conductors are calculated so that the impedance of the line remains everywhere approximately 50 Ω . At the end of the coaxial line, a 50 Ω load absorbs the r.f. wave. Therefore the r.f. wave interacting with the atoms is a travelling one and the r.f. magnetic field strength seen by the atoms depends only on the r.f. power delivered by the generator (no resonance effects due to the length of the coaxial line). Moreover, since the r.f. travelling wave propagates perpendicularly to the atomic beam, any Doppler shift of the resonances is avoided.

The r.f. wave is produced by a commercial r.f. synthesizer (Rhode & Schwartz SMS 0.4-

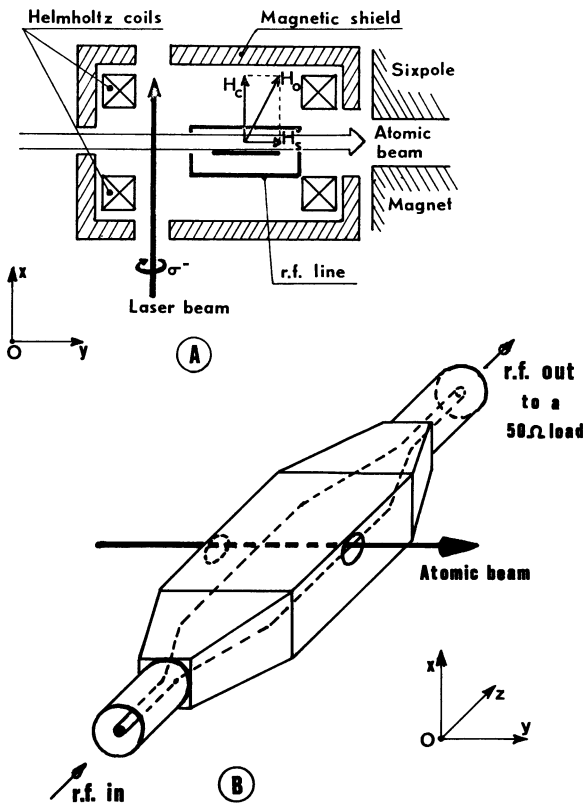


Fig. 2. — Schematic view of the interaction region (a) and of the r.f. line (b).

1 040 MHz) followed by a frequency doubler and a linear wide band amplifier (2 W, 0.4-2 GHz). A 2 W r.f. power, continuously tunable up to 2 GHz, is then available.

Two Helmholtz coils produce a static magnetic field H_c parallel to the $x'x$ direction of the laser beam. H_c is roughly homogeneous over the interaction zones with the laser field and the r.f. field.

A magnetic shield of Armco iron and mu-metal sheets surrounds the whole interaction zone. In order to avoid non adiabatic transitions for the optically pumped atoms one has to preserve a weak magnetic stray field from the sixpole magnet [12]. For this reason, a sufficiently large hole is bored through the magnetic shield in front of the entrance of the sixpole magnet. This stray field H_s points approximately parallel to the atomic beam ($y'y$ direction) ; its amplitude over the r.f. coaxial line is relatively weak (typically 0.4 G) and strongly inhomogeneous.

4. Observed resonances and line shapes.

The observable resonances depend on the relative orientation of the static and r.f. magnetic fields seen by the atoms. The total static magnetic field is $H_0 = H_c + H_s$. When the d.c. current in the Helmholtz coils is high enough to get $H_c \gg H_s$ the static field is roughly homogeneous and parallel to the

$x'x$ direction (case a). When the coils are not energized the total static field is strongly inhomogeneous and parallel to the $y'y$ direction (case b). Let H' be the r.f. magnetic field seen by the atoms. In a first approximation we can consider that H' is parallel to the $y'y$ direction and the induced r.f. magnetic transitions are the $\Delta m_F = \pm 1$ (respectively $\Delta m_F = 0$) for case a (respectively case b).

If we enter into more details, since the r.f. magnetic field lines turn around the internal conductor, the r.f. field H' seen by the atoms exhibits two components H'_y and H'_x which are different from zero (see Fig. 3). The first one is an even function of the position of the atoms, the second one an odd function.

As explained above H'_y will induce some magnetic transitions ($\Delta m_F = \pm 1$ in case a or $\Delta m_F = 0$ in case b) but H'_x will induce the other ones ($\Delta m_F = 0$ in case a and $\Delta m_F = \pm 1$ in case b) ; this requires, of course, a higher r.f. power due to the low amplitude of H'_x compared to H'_y .

Moreover, it is clear in figure 3 that H'_x is confined in two separated zones at the entrance and the exit of the r.f. line on the atomic path.

The atoms of the beam interact successively with the r.f. magnetic field of these two zones : this geometry is similar to the one used in Ramsey fringes technique [13]. But one has to notice that in our case the sign of H'_x changes between the two zones corresponding to a difference in phase of π for the two r.f. fields successively seen by the atoms. For this reason one expects that the resonance curve will present a minimum at the exact resonance frequency.

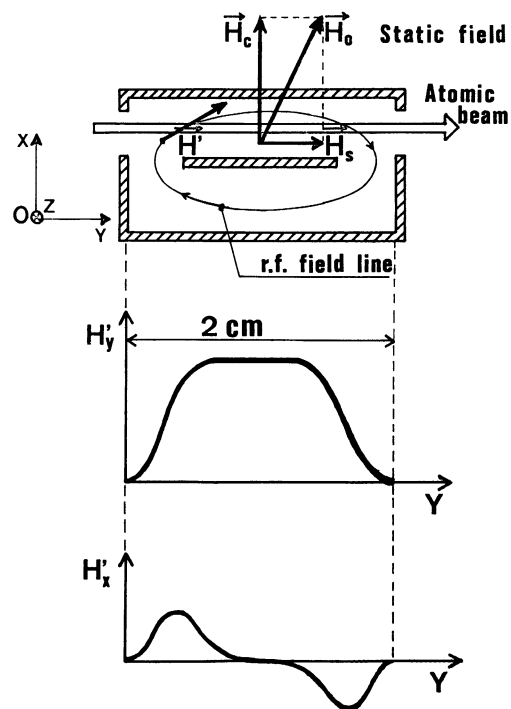


Fig. 3. — Geometry and relative orientation of the static (H) and the r.f. (H') magnetic fields.

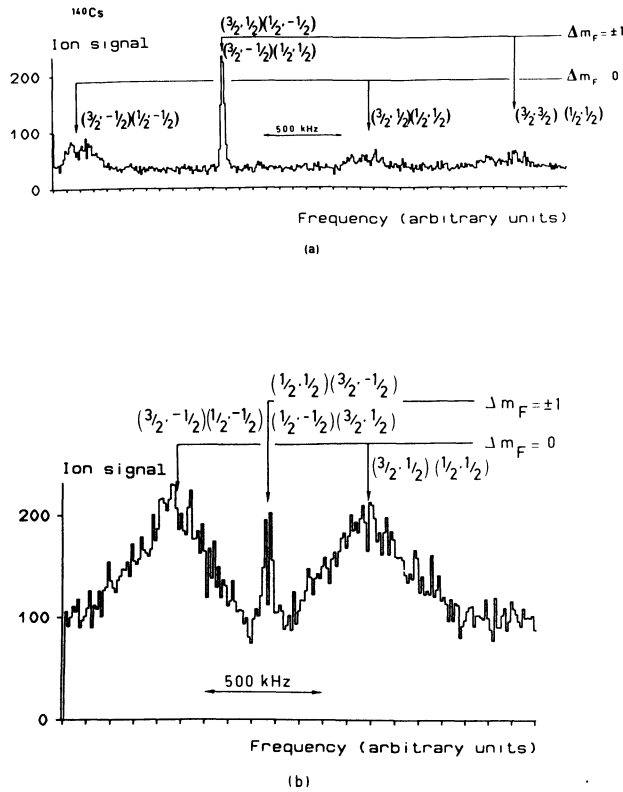


Fig. 4. — Zeeman splitting of the $\Delta F = 1$ hyperfine transition for ^{140}Cs . Recordings a and b have been obtained for $H_c \gg H_s$ (case a) and $H_c = 0$ (case b) (see text). For both recordings the frequency scale is 10 kHz/step.

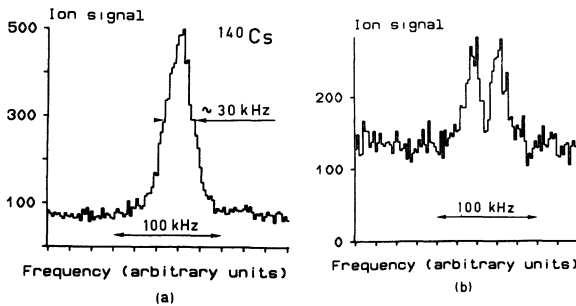


Fig. 5. — High resolution recordings (frequency step: 2 kHz) of the central resonance curve corresponding to the first order field independent unresolved doublet for ^{140}Cs . As in figure 3 recordings a and b correspond respectively to case a and b for the static field (see text). Notice, in the b recording, the strong dip at the center of the resonance.

Figures 4 and 5 present the experimental curves obtained for ^{140}Cs ($I = 1$) with a static field corresponding either to cases a or b. The expected structures have been observed. The whole Zeeman splitting of the $\Delta F = 1$ hyperfine transition has been first recorded at relatively low resolution with a frequency step of 10 kHz (Figs. 4a and b). The relative intensities of the $\Delta m_F = 0$ and $\Delta m_F = \pm 1$

components are coherent with the explanations given above. The resonances the frequency of which are first order field dependent are broadened due to the inhomogeneities of the static field particularly in case b. For these transitions the line shapes are linked in a complicated way to the field inhomogeneities [13], and no attempt has been done to analyse them precisely.

On the other hand, one resonance is only second order field dependent. For the case of ^{140}Cs (even isotope), it is an unresolved doublet of two $\Delta m_F = \pm 1$ transitions

$$(I + 1/2, 1/2) \leftrightarrow (I - 1/2, -1/2)$$

and

$$(I + 1/2, -1/2) \leftrightarrow (I - 1/2, +1/2).$$

For this resonance, recordings obtained at high resolution with a frequency step of 2 kHz are displayed in the figure 5a and b. The frequency width of the first recording (30 kHz) is not far from the limit value deduced from the interaction time (~ 20 kHz). The second recording, obtained in case b, exhibits a strong dip, as expected, just at the center of the resonance.

Finally an important remark may be made on the geometry of the set-up: the r.f. field \mathbf{H}' seen by the atoms does not turn around the static field \mathbf{H}_0 , it remains in the xy plane (see Fig. 3). This eliminates the possibility of a systematic error in the determination of the resonance frequency due to the Millmann effect [13, 14].

5. Experimental procedure.

In order to determine the hyperfine splitting of the ground state with a high accuracy the following steps have been successively performed.

An approximate value of the hyperfine structure is first given by laser spectroscopy to within an accuracy of a few MHz. Then the static field is set to $\mathbf{H}_0 = \mathbf{H}_s$ (no current in the coils) and the r.f. experiment is performed. Since the resonance are broadened by the field inhomogeneities the r.f. frequency increment may be chosen relatively large (typically 20 kHz) which is time saving.

When this low resolution spectrum of the Zeeman splitting is obtained, the field independent component is searched in the center of this splitting. High resolution spectra (typically 1 kHz increment) of this resonance are recorded for one or several values of H_0 . One has to choose $\mathbf{H}_0 \approx \mathbf{H}_s$ (case b) for odd isotopes since the involved transition is a $\Delta m_F = 0$ one, and $H_0 \gg H_s$ (case a) for even isotopes ($\Delta m_F = \pm 1$ doublet). In order to get the hyperfine structure in zero field from the measured frequency of the transition one has to take into account the second order correction calculated from the usual Breit-Rabi formula [13]. The static field H_0 is known either from the current in the coils owing to a previous calibration, or from a recording of the

whole Zeeman splitting in the same field H_0 [9]. In any case this correction is always small (less than 20 kHz).

6. Résultats and discussion.

According to the frequency limitation of our r.f. set up the experiment has been carried out only for ^{76}Rb , ^{77}Rb , ^{82}Rb , $^{84}\text{Rb}^m$, ^{140}Cs and ^{226}Fr , for which the hyperfine structures are smaller than 2 GHz.

Taking into account the second order correction, the deduced values of the hyperfine structures are gathered in table I. For ^{140}Cs , $^{84}\text{Rb}^m$ two independent measurements have been done with different static fields and the results differ by less than 1 kHz. We therefore adopted a 2 kHz error bar. For ^{76}Rb , ^{77}Rb , ^{82}Rb and ^{226}Fr only one high resolution spectrum has been recorded. The quoted uncertainty is increased up to 3 kHz in the case of ^{82}Rb and ^{226}Fr and up to 4 kHz in the case of ^{76}Rb and ^{77}Rb for which the resonances are slightly broader (~ 50 kHz) and noisier.

Previous results obtained by laser spectroscopy are also collected in table I. The very precise values obtained here are always coherent with these results from optical spectroscopy. This is a convincing test of the reliability of the error bars quoted in our previous laser spectroscopy experiments [4-6].

An estimate of the sensitivity of the set-up has been given in a previous paper [9]. The results obtained here allow us to be more explicit about this point. A recording of the resonance curve obtained for ^{76}Rb is displayed in figure 6. We are clearly far above the limit of sensitivity. The production yield

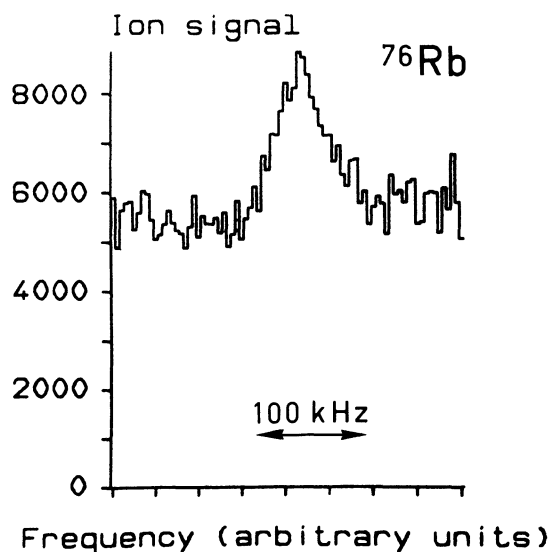


Fig. 6. — High resolution recording of the central resonance obtained for the least abundant isotope studied, ^{76}Rb , (frequency scale : 4 kHz/step).

for this isotope being 2×10^6 atoms s^{-1} [4] the limit of sensitivity is certainly below 10^6 atoms s^{-1} . The sensitivity of the present set-up is therefore higher than the best one obtained in classical ABMR technique [15]. In conclusion, it is important to emphasize that as far as nuclear magnetic moments could be very precisely determined in the future for long radioactive sequences, the present method, owing to its extreme accuracy and sensitivity, would be fully adapted in determining hyperfine anomalies.

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