Direct Measurement of Nuclear Magnetic Moments of Radium Isotopes

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The nuclear magnetic moments of ^{213}Ra and ^{225}Ra have been measured at ISOLDE (isotope separator at the CERN synchrocyclotron) by observation of the Larmor precession of optically pumped atoms in a fast beam. The results $\mu_I(^{213}\mathrm{Ra}) = 0.6133(18)\mu_N$ and $\mu_I(^{225}\mathrm{Ra}) = -0.7338(15)\mu_N$ provide an accurate test of *ab initio* and semiempirical calculations from optical hyperfine structures in Ra I and Ra II.

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Over the past few years collinear fast-beam–laser spectroscopy on line with accelerators has proved to be a very fruitful technique for the study of exotic very unstable nuclei.[1] Nuclear magnetic moments, for instance, are deduced from hyperfine-structure (hfs) measurements through the magnetic interaction constant A and the nuclear spin I. This requires of course, the knowledge either of the magnetic field

$$H_e(0) = AIJ/\mu_I \quad (1)$$

created by the valence electrons at the nucleus, or of the nuclear magnetic moment of at least one isotope. Moreover, this procedure implies that the hyperfine anomaly is negligible, and consequently the uncertainty on the nuclear results is rather large (1% typically) even if the hfs constants are known to a much better precision.

Such measurements in a series of Ra isotopes were reported three years ago.[2] Since no direct measurement of the nuclear magnetic moment had been performed previously for any of the Ra isotopes, it was necessary to evaluate $H_e(0)$, and this is the main source of the estimated $\pm 5\%$ error in the μ_I values. There have been attempts to reduce this error by both theoretical *ab initio* calculations[3-5] and improved semiempirical analyses.[6]

We report here the first direct measurement of the nuclear magnetic moments of ^{213,225}Ra which eliminates the uncertainty about $H_e(0)$ in the determination of the μ_I values for the whole isotopic sequence and also tests the atomic hfs theory.

The experimental method consists of our measuring the Larmor precession of the ground-state magnetic moment in an external static magnetic field by observing the fluorescence of a fast atomic beam interacting with a collinear resonant laser beam. Such experiments were first performed with paramagnetic ($J \neq 0$) neutral atoms[7] or ions.[8] When the experiment is performed with diamagnetic atoms ($J = 0$ ground state), the gyromagnetic factor g_I and thus the nuclear magnetic moment μ_I can be deduced directly from the precession frequency.

The principle of the method has been described in a previous paper devoted to a pilot experiment on Ba isotopes[9]: The fast neutral atoms are first aligned (or oriented) by optical pumping through linearly (or circularly) polarized laser excitation (zone A in Fig. 1). Then, these aligned (or oriented) atoms precess at the Larmor frequency in a strong external magnetic field (zone B). In this region, the atoms are out of resonance with the laser because of the large Zeeman splitting of the upper level of the transition. At the exit of the magnet gap the fast atoms are reexcited by the laser light and the fluorescence intensity is strongly dependent on the Larmor rotation experienced by the magnetic moments. This results in a fringe structure of the fluorescence signal which is recorded as a function of the static magnetic field. The g_I factor of the atom is readily deduced from the period of the fringes, provided that the atomic velocity, the magnetic field amplitude, and the interaction length with the magnetic field are known. Since this last quantity is difficult to determine precisely, we adopt a comparative method and use as a calibration the Larmor precession of another nuclide for which g_I is known (^{135,137}Ba in our case).

The accuracy of the method increases with the number of observable fringes. Thus, for nuclei with $I > \frac{1}{2}$, it is preferable to use alignment rather than orientation since alignment doubles the number of fringes to be recorded.[7] The number of expected fringes can be estimated from the preliminary g_I values available in Ref. 2. It turns

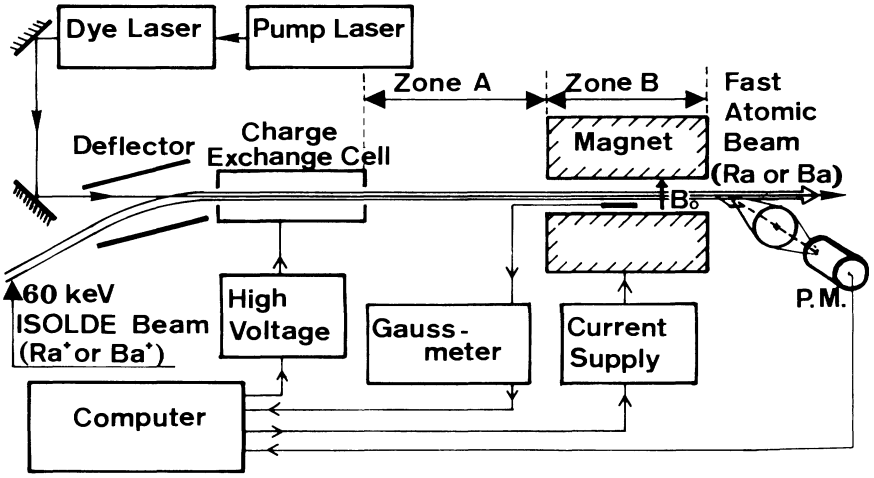

FIG. 1. The experimental setup, including a frequency-stabilized cw dye laser system, deflection of the ion beam into the laser beam axis, and neutralization cell. The fluorescence is detected downstream from the optical pumping zone (A) and the magnet (B). The observation direction is perpendicular to the magnetic field direction.

out that the most favorable isotopes are ^{213}Ra ($I=\frac{1}{2}$) and ^{225}Ra ($I=\frac{1}{2}$).

The experiment has been carried out at the ISOLDE mass separator[10] at CERN. The radium isotopes are produced by spallation of ^{232}Th in a ThC_2 target with 600-MeV protons from the CERN synchrocyclotron. The reaction products are ionized, accelerated up to 60 keV, and mass separated. The yields are about 10^7 ions/s for ^{213}Ra and 10^9 ions/s for ^{225}Ra. The same source also delivers the beam of stable Ba isotopes needed for calibration.

The 60-keV ion beam is transmitted to the collinear laser-spectroscopy apparatus (see Fig. 1). This is a modified version of the standard setup for neutralized fast beams.[11] The ion beam passes through a sodium vapor cell which is heated to 250 °C to convert 70% of the ions into neutral atoms by near-resonant charge exchange. Downstream from the charge-exchange cell, a free flight path of 60 cm (zone A) ensures efficient optical pumping and the 40-cm-long pole gap of an electromagnet defines the region of nuclear Larmor precession. The magnetic field B_0 can be varied from 0 up to 9500 G; it is measured with two calibrated Hall probes. The fluorescence at 90° with respect to the magnetic field axis is then detected in a field-free region of 20 cm. Two dye laser systems operate simultaneously, on the resonance lines of Ra I ($7s^2\,{}^1S_0 \rightarrow 7s7p\,{}^1P_1$) at 482.6 nm and Ba I ($6s^2\,{}^1S_0 \rightarrow 6s6p\,{}^1P_1$) at 553.6 nm. Doppler tuning is used to set the laser frequency to one of the hyperfine components: For $I=\frac{1}{2}$ the strongest contrast in the fringe pattern is observed by the excitation of the $F=I$ level of the upper state, whereas for $I\geq\frac{3}{2}$ the $F=I-1$ level is preferable. To record the fringes, the current supply of the magnet is scanned slowly and linearly, and the fluorescence signal is stored in a microcomputer together with the output signal from the gaussmeter. Before each slow scan, the electromagnet is carefully demagnetized. This gives the fluorescence signal of ^{225}Ra shown in Fig. 2(a).

Since the slow recording procedure suffers from the fluctuations of the proton beam intensity, we tended to use a fast scanning mode: The magnet current is continuously swept up and down (30 ms per channel) over the last few fringes, and the fluorescence signals of successive scans are added in the computer. Typical recordings of the fluorescence and gaussmeter signals are displayed in Figs. 2(b) and 2(c). The strong hysteresis visible in Fig. 2(c) indicates that the scanning procedure must be precisely defined. To prevent systematic errors, the following precautions were taken: (i) The speed and the amplitude were kept the same for all the fast cyclic scans, including those on the reference nuclides. (ii) The magnet current experienced a few dead cycles before the data acquisition was started, to ensure a stable hysteresis loop during the effective recording. (iii) In the data-analysis procedure, only the spectra recorded in the same way were compared among themselves.

Following the calculations of Ref. 9, the phase of the oscillating signal for circularly polarized pumping light is

$$\Phi=(g_I\mu_N B_0/\hbar v)\int f(y)dy, \qquad (2)$$

where v is the velocity of the atoms and $B_0 f(y)$ is the magnetic field at position y along the beam path, B_0 being the maximum field in the magnet gap where $f(y)=1$ (a factor of 2 must be added if one is dealing with aligned rather than with oriented atoms). The equivalent length $K=\int f(y)dy$ cancels out in relative measurements. The velocity of the atoms is known with an accuracy of 5×10^{-5}.[11] The stability of the Hall probe has been found to be a few parts in 10^4 over 2 h.

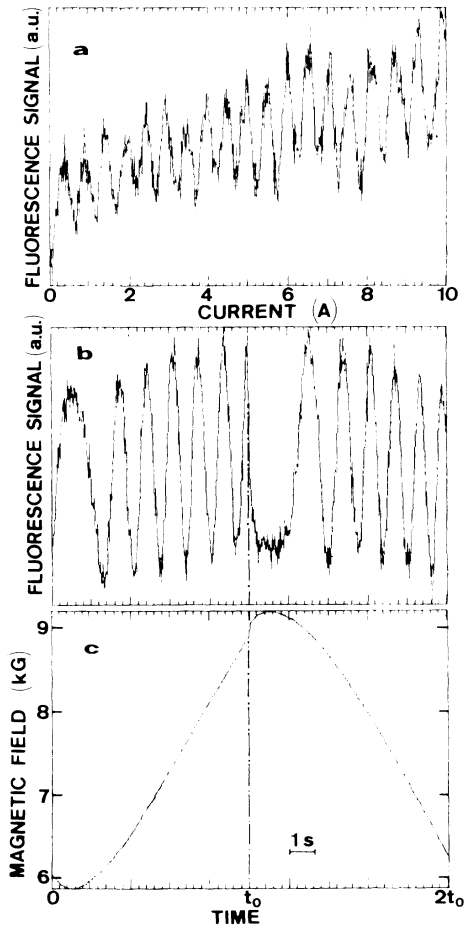

FIG. 2. (a) Fluorescence signal obtained with ^{225}Ra as a function of the magnet current (slow recording procedure). (b) and (c) Fluorescence of ^{225}Ra and Gaussmeter signals as functions of time (fast cyclic recording procedure). The magnet current increases linearly from 5 to 10 A between 0 and t_0 and decreases back from 10 to 5 A between t_0 and $2t_0$. The discontinuities which occur at t_0 and $2t_0$ are due to a short fixed dead time occurring each time the current slope is reversed; it is not shown in the figure since the horizontal scale corresponds to the counting time only. The recording is obtained with an integration time of 900 ms per channel (30 scans) in 500 channels.

During the run, reference spectra were recorded periodically so that the radium and the reference spectra to be used for comparison in the data analysis were taken at relatively short time intervals. This procedure also averages the possible error due to imperfect setting of the collinear atomic and laser beams.

The fringe pattern has been analyzed by use of a Fourier-transform program. In addition, a few recordings were treated with the direct fitting procedure described in Ref. 9. Both methods, applied to the same spectrum, give identical oscillation frequencies.

TABLE I. Measured (^{213}Ra and ^{225}Ra) and deduced (other isotopes) nuclear magnetic moments (in units of μ_N). For the latter, the hfs of Refs. 6 and 2 have been used.

Isotope	μ_I	Isotope	μ_I
209	0.865(13)	223	0.2705(19)
211	0.8780(38)	225	−0.7338(15)
213	0.6133(18)	227	−0.4038(24)
221	−0.1799(17)	229	0.5025(27)

The results are compiled in Table I. If we compare the spectra of ^{213}Ra and ^{225}Ra with those of ^{137}Ba, and apply the diamagnetic correction according to Lederer and Shirley,[12] the nuclear moments of these two isotopes are obtained from $\mu_I(^{137}\text{Ba}) = 0.937\,365(20)\mu_N$.[12,13] Their errors reflect the standard deviation of the experimental results. As a test of the reliability of the method, spectra of ^{135}Ba have also been recorded. The measured ratio $\mu_I(^{137}\text{Ba})/\mu_I(^{135}\text{Ba}) = 1.1202(22)$ is consistent with the known value 1.11865(3).[13] The new directly measured radium moments are fairly close to those previously determined indirectly; thus the nuclear physics discussion presented in Ref. 2 and the conclusions about octupole-deformed nuclear ground state[14,15] remain valid. The μ_I values of the other isotopes are deduced from the directly measured ones by use of the ratios of the hfs constants $A(^2P_{1/2})$ which are essentially free of hfs anomaly effects.[2,6] The value of $\mu_I(^{209}\text{Ra})$ has been taken from the hfs of the $7s7p\ ^1P_1$ state in Ra I,[6] and includes an estimated 1% error for the hfs anomaly.

If we directly compare spectra of ^{225}Ra and ^{213}Ra, the ratio $^{225}g_I/^{213}g_I$ is found to be −1.1981(36). This value allows us to recalculate the hyperfine anomaly between these two isotopes in the $^2S_{1/2}$ ionic ground state from the known hfs constants. We find

$$
\begin{aligned}
&{}^{225}\Delta^{213}(^2S_{1/2}) \\
&\quad = [^{225}A(^2S_{1/2})\,{}^{213}g_I/{}^{213}A(^2S_{1/2})\,{}^{225}g_I] - 1 \\
&\quad = +0.008(4), \qquad (3)
\end{aligned}
$$

in agreement with the result $^{225}\Delta^{213} = +0.004(3)$ quoted in Ref. 2.

The direct g_I-factor measurement now provides a crucial test of the recent theoretical *ab initio* calculations and semiempirical evaluations of the magnetic hyperfine fields $H_e(0)$ in the $7s\ ^2S_{1/2}$ and $7p\ ^2P_{1/2}$ states of Ra II. This comparison is given in Table II, where the calculated values of $H_e(0)$ and the corresponding magnetic moments from the hfs of ^{213}Ra are listed together with the direct experimental results.

For the $7s\ ^2S_{1/2}$ state, it turns out that both the theoretical[3-5] and the recent refined semiempirical[6] analyses systematically overestimate $H_e(0)$ by about 3%–6%, while the rather good agreement with the earlier semiempirical values[2] following Kopfermann[16] is

TABLE II. Comparison of the hyperfine fields $H_e(0)$ from *ab initio* and semiempirical calculations with the direct experimental results. The semiempirical values for $7p\ ^2P_{1/2}$ and the $7s7p$ configuration of Ra I (Ref. 6) are too inaccurate to be considered here.

Atomic state (Ra II)	$H_e(0)$ (T)	$\mu_I(213)$ (μ_N)	Method	Ref.
$7s\ ^2S_{1/2}$	1226(4)	0.6133(18)	Expt.	This work
	1210(60)	0.62(3)	GFS[a]	2
	1285(65)	0.585(30)	GFS[a]	6
	1293(40)	0.58(2)	RMBPT[b]	3
	1304(65)	0.58(3)	RMBPT[b]	4
	1266(13)	0.594(6)	RMBPT[b]	5
$7p\ ^2P_{1/2}$	242.0(10)	0.6133(18)	Expt.	This work
	246(12)	0.60(3)	RMBPT[b]	4
	243(3)	0.612(8)	RMBPT[b]	5

[a]Semiempirical values with use of the Goudsmit-Fermi-Segrè formula with different corrections (here the 5% errors correspond to the general reliability of the procedure and do not mean that both values are consistent).

[b]Relativistic many-body perturbation theory (errors as estimated by the authors).

probably fortuitous. As an accuracy of 1% is claimed for the atomic theory, one might query the adequacy of the relatively crude assumptions for the nuclear magnetization distribution. This would explain why the calculated fields and moments for the $7s\ ^2P_{1/2}$ hyperfine structure are closer to the experimental values.

We have shown that the nuclear Larmor precession method on atoms with $J=0$ ground states is capable of providing precise magnetic moments. At present it is just accurate enough for the observation of hyperfine anomalies. In order to measure them to some precision throughout the isotopic sequence, it would be sufficient to reduce the error by 1 order of magnitude. For that purpose, the length of the magnet can be considerably increased, but only a slight increase seems possible for the maximum field strength. It should also be possible for one to observe the fringes without changing the field amplitude, e.g., by synchronously scanning the atomic beam velocity and the laser frequency. With such improvements a gain in precision by a factor of 10 may be attainable.

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