Nuclear Orientation of Radon Isotopes by Spin-Exchange Optical Pumping

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This paper reports the first demonstration of nuclear orientation of radon atoms. The method employed was spin exchange with potassium atoms polarized by optical pumping. The radon isotopes were produced at the ISOLDE isotope separator of CERN. The nuclear alignment of ^{209}Rn and ^{223}Rn has been measured by observation of γ-ray anisotropies and the magnetic dipole moment for ^{209}Rn has been measured by the nuclear-magnetic-resonance method to be $|\mu| = 0.838\,81(39)\mu_N$.

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Nuclear polarization methods have been very useful for studies of nuclear structure and for investigations of fundamental nuclear processes. For example, the polarization of noble-gas atoms has been especially useful for studies of time-reversal invariance[1] and nuclear electric dipole moments (EDM's).[2] This special feature of the noble-gas atoms is due to the long spin-depolarization times. One might also expect that polarized radon atoms would be very useful for such studies, and in particular for measurements of nuclear EDM's,[3] because atomic electron screening effects are reduced as the atomic number increases and an enhancement of nuclear EDM by state mixing is expected for some radon isotopes. But to date no method has been available for the production of nuclear orientation of gaseous radon.

We report here the first demonstration of nuclear orientation of radon. The radon nuclei are oriented by spin-exchange collisions with alkali atoms that are polarized by optical pumping. This method has been employed previously to polarize the lighter noble-gas atoms and most extensively xenon for which polarizations of $\gtrsim 70\%$ are readily achieved.[4-6]

The cross section for spin exchange is larger for the heavier noble-gas atoms because of, in part, the importance of the formation of alkali–noble-gas-atom molecules.[7,8] For this reason we expected that radon would be readily polarized by spin exchange on alkali atoms too. A detailed discussion of the spin-exchange process is given by Happer *et al.*[6] On the other hand, for radon we anticipated significantly larger depolarization because of the wall sticking compared to xenon. So special attention has been paid to reduce the quadrupole relaxation on the cell wall as described later.

As in earlier studies of radioactive xenon isotopes,[4,5] we have used the γ-ray anisotropy method to observe the nuclear orientation. We chose ^{209}Rn (half-life $t_{1/2} = 29$ min) for these studies as it has a reasonably well-established decay scheme, enabling us to predict the expected γ-ray anisotropy, and because its hyperfine structure has recently been studied by collinear laser spectroscopy.[9] The laser spectroscopy has also established the spin $I = \frac{5}{2}$ by direct measurement and a positive sign of the magnetic moment of ^{209}Rn. Our direct measurement of the magnetic moment would provide a calibration of the hyperfine magnetic field of the radon atom.

We also polarized ^{223}Rn [$t_{1/2} = 23$ min (see Borge *et al.*[10])] by the spin-exchange method and the γ-ray anisotropies of several γ transitions have been measured. The decay scheme of this isotope has not been established yet, but a preliminary study of the γ-ray lines and multipolarities has been published.[10]

The radon activity was produced at the ISOLDE isotope-separator facility of CERN by spallation reaction of 600-MeV protons on a 55-g/cm^2 ThC_2 target.[11] The optical-pumping cells were filled with radon isotopes with the apparatus illustrated in Fig. 1(a). The mass-separated 60-keV Rn^+ was stopped in a thin (7.6 μm) tantalum foil within a small target chamber evacuated to pressures of $\simeq 10^{-7}$ Torr. The target chamber has lines to gas bottles containing hydrogen and nitrogen, a sidearm containing potassium as a getter, and a port to a valve on which the cell glassware is attached.

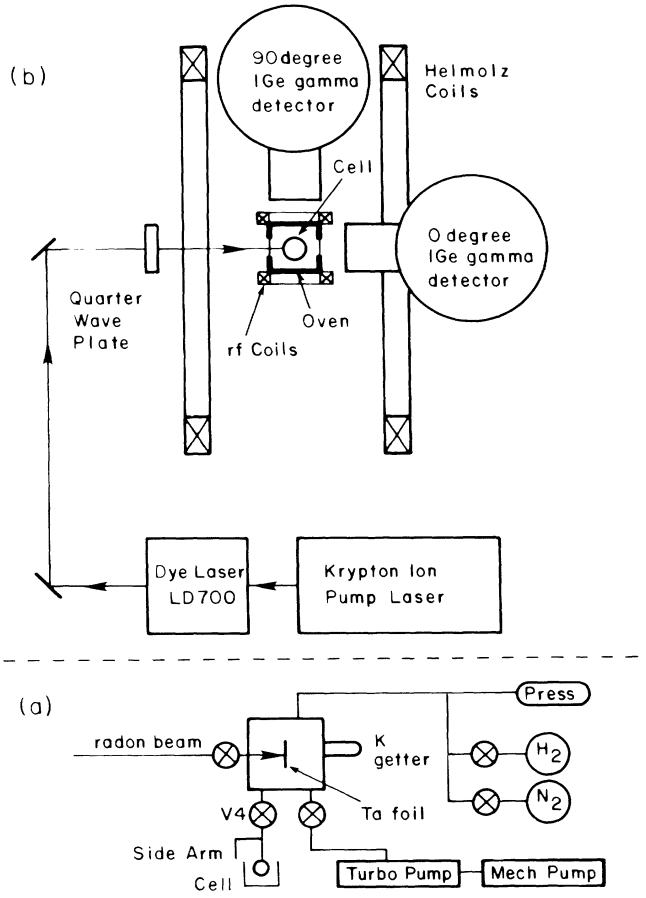

FIG. 1. (a) Cell-filling and (b) optical-pumping apparatus.

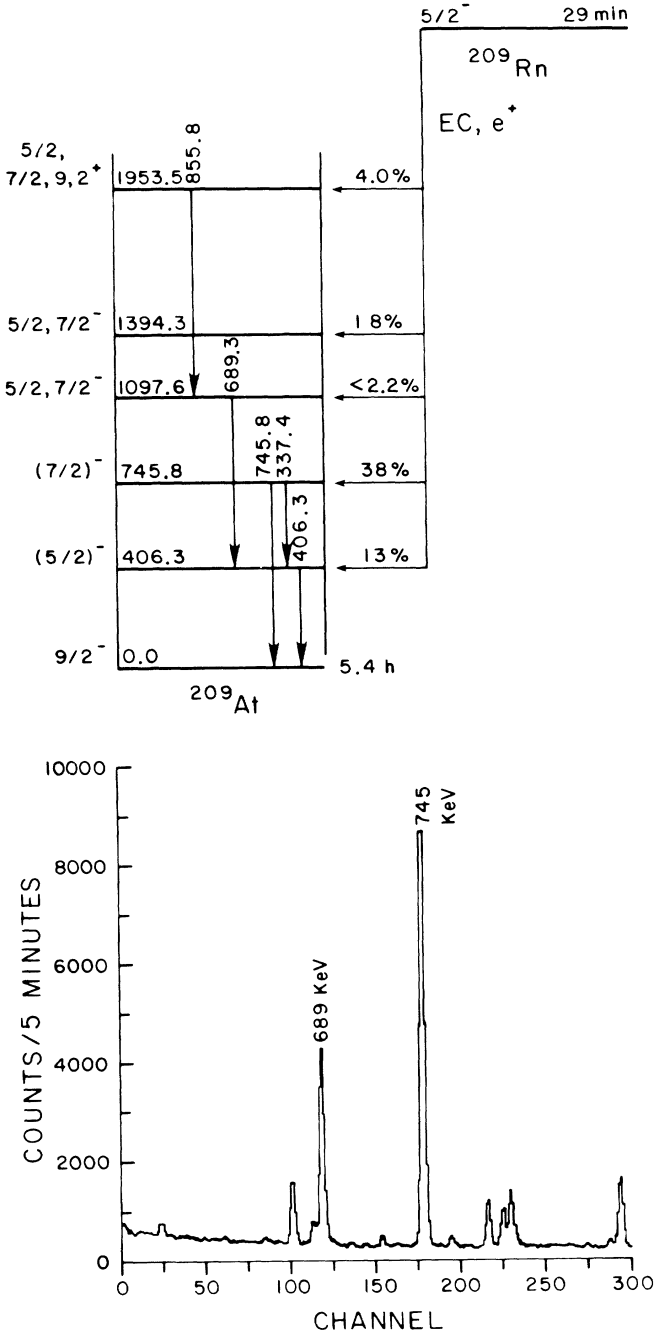

FIG. 2. γ-ray spectrum of ^{209}At following ^{209}Rn decay. A simplified decay scheme of ^{209}Rn is also shown.

In advance of the run, we prepared several Pyrex glass cells (1 cm diam) loaded with a few milligrams of potassium and ~5 Torr of hydrogen gas. Potassium was used as the alkali for spin exchange, rather than rubidium as in past xenon experiments,[4,5] to permit operation of the cells at a high temperature (250 °C instead of 190 °C and thereby reduce depolarization of radon by wall sticking. Hydrogen gas was introduced and the cells were heated for a few days at 80 °C to form a surface layer of potassium hydride which also reduces wall relaxation.[12]

After a few seconds of implantation, the radon beam is turned off, the chamber is isolated, the radon is driven from the foil by resistive heating, and the gaseous radon is then condensed in the cell by liquid-nitrogen cooling. Gaseous nitrogen (400 Torr) and hydrogen (10 Torr) are added and the cell is pulled off.

The cell containing radon is placed in an oven and heated by flowing hot air to about 230 °C [see Fig. 1(b)]. A krypton-ion laser (Coherent K3000) and a dye laser (Coherent CR699), operating in standing-wave mode with LD700 dye, produces 0.8 W of light at the 769.9-nm D_1 line of potassium at the cell. The light is right- or left-circularly polarized with a $\lambda/4$ plate placed just in front of the cell. A few gauss of magnetic field parallel to the light beam is produced by a single pair of Helmholtz coils and a power supply stable to $1/10^4$.

The nuclear alignment of the radon was monitored by observation of the γ-ray anisotropy between two IGe detectors (12%, Ortec) placed at 0° and 90° relative to the magnetic field. The γ-ray spectra were acquired for typically 5-min intervals with a two-channel CAMAC analog-to-digital converter and histogramming memory system that was interfaced to an Apple IIe microcomput-

TABLE I. Summary of anisotropies for strong ^{209}At γ lines following ^{209}Rn decay.

E_γ (keV)	Spin sequence	Anisotropy R	$R-1$ (%)
337	$(\frac{7}{2}^-)$-$(\frac{5}{2}^-)$	0.903(14)	-9.7 ± 1.4
408	$(\frac{5}{2}^-)$-$\frac{9}{2}^-$	1.009(7)	$+0.9\pm0.7$
689	$\frac{5}{2}, \frac{7}{2}^-$-$\frac{5}{2}^-$	1.079(22)	$+7.9\pm2.2$
745	$(\frac{7}{2}^-)$-$\frac{9}{2}^-$	1.129(14)	$+12.9\pm1.4$

er. An acquisition program was used to calculate γ anisotropies on line.

The γ-ray spectrum for ^{209}Rn in the vicinity of the 745-keV line is illustrated in Fig. 2. A simplified decay scheme of ^{209}Rn, illustrating some of the prominent γ-ray transitions is also shown in Fig. 2. The anisotropy ratios for the strong γ-ray transitions are given in Table I. The quantity R is the ratio of counts in the 0° detector to counts in the 90° detector for "laser on" divided by the same ratio for "laser off." From these results we see that the ^{209}Rn is definitely aligned. The anisotropy of the 745-keV line is predicted to be 1.3 on the assumption that it is a pure $M1$ transition and that the spin for the 745-keV level is $\frac{7}{2}$ and that all initial ^{209}Rn nuclei with spin $\frac{5}{2}$ are in the $m=+\frac{5}{2}$ magnetic substate. The observed anisotropy indicates that the alignment is about 50% of the maximum possible alignment.

To observe nuclear alignment of ^{223}Rn, a cell containing ^{223}Rn was prepared in the same way as described above for the ^{209}Rn cell. The anisotropies for prominent γ-ray lines are given in Table II. Several of the γ-ray lines exhibit a significant anisotropy. A complete analysis of the γ anisotropies for both isotopes ^{209,223}Rn will be presented in a future publication; these data will be useful for clarification of the decay schemes.

The magnetic moment of ^{209}Rn, relative to the accurately known moment of ^{129}Xe^m,[4,5] was measured by a search for resonant destruction of nuclear alignment induced by an oscillating magnetic field. The resonance data are shown in Fig. 3. The two resonance peaks, power broadened to facilitate the search, correspond to

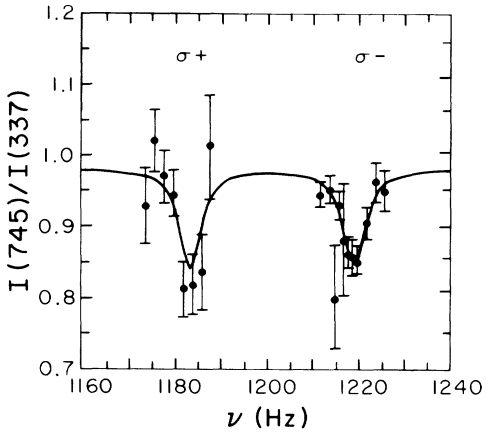

FIG. 3. The NMR resonance curves for ^{209}Rn. Count ratios between 745- and 337-keV lines for the 0° detector are plotted. As seen in Table I, these two lines show opposite sense of anisotropy. The two resonances correspond to the $\sigma\pm$ pumping (see text).

opposite circular polarizations of the laser light; the frequency shift arises from the collisional field $B_{\rm coll}$ because of spin exchange with the polarized alkali.[7] The measurements on ^{209}Rn and ^{129}Xe^m were performed in separate, but similar, cells. The centroid frequencies of the NMR resonances for these isotopes are given in Table III, along with the diamagnetic corrections and the magnetic moment of ^{208}Rn determined from the relationship

$$|\mu^{\rm Rn}| = |\mu^{\rm Xe}| \frac{(\nu_+^{\rm Rn}+\nu_-^{\rm Rn})I^{\rm Rn}(1-\sigma^{\rm Xe})}{(\nu_+^{\rm Xe}+\nu_-^{\rm Xe})I^{\rm Xe}(1-\sigma^{\rm Rn})},$$

where the spins I are $\frac{11}{2}$ and $\frac{5}{2}$ for ^{129}Xe^m and ^{209}Rn, respectively, and the diamagnetic corrections $1/(1-\sigma)$ are taken from Ref. 13.

In conclusion, the present work has demonstrated the feasibility of the polarization of radon nuclei by the spin-exchange optical-pumping method. The spin transfer rate is apparently faster for radon than for xe-

TABLE II. γ-ray anisotropies of ^{223}Fr from the decay of oriented ^{223}Rn.

E_γ (keV)	Anisotropy R	$R-1$ (%)
159.9	0.958(18)	-4.2 ± 1.8
171.8	1.082(24)	$+8.2\pm2.4$
206.3	0.990(25)	-1.0 ± 2.5
416.0	0.933(14)	-6.7 ± 1.4
491.4	1.092(39)	$+9.2\pm3.9$
591.8	1.014(12)	$+1.4\pm1.2$
621.5	0.936(40)	-6.4 ± 4.0
635.2	1.011(15)	$+1.1\pm1.5$
723.2	1.054(33)	$+5.4\pm3.3$

TABLE III. Frequencies of NMR resonances for ^{209}Rn and ^{129}Xe^m and related quantities.

	^{129}Xe^m	^{209}Rn
I	$\frac{11}{2}$	$\frac{5}{2}$
ν_+ (Hz)	586.750(5)	1184.66(69)
ν_- (Hz)	588.292(60)	1218.77(32)
$\nu_- - \nu_+$ (Hz)	1.542	34.11
Collisional field $B_{\rm coll}$ (G)	0.0063	0.067
External field B_0 (G)	4.790	4.790
Diamagnetic correction[a] $1/(1-\sigma)$	1.007092	1.0195
Magnetic moment[b] $\lvert\mu\rvert$ (μ_N)	0.891223[c]	0.83881(39)

[a] Reference 13.
[b] Corrected for diamagnetism.
[c] Reference 5.

non, as evidenced by the larger frequency shift $\nu_- - \nu_+$ in Table III. In addition, we have determined the magnetic moment of ^{209}Rn by direct measurement. This result in combination with the measurement of the hyperfine structure of atomic transitions in ^{209}Rn by collinear laser spectroscopy determines the magnetic hyperfine field (to be published), and, apart from hyperfine anomaly effects, also the magnetic moments of the other radon isotopes whose hyperfine structure has been measured.[9] Here we note that our value of the magnetic moment 0.83881 agrees fairly well with the estimate 0.82 given in Ref. 9. The γ anisotropies of ^{209}Rn and ^{223}Rn, to be published, will aid in the making of spin-parity assignments of nuclear levels. This first successful alignment of radon suggests that measurements of the nuclear EDM may be possible by high-resolution NMR experiments; the sensitivity of such a measurement is presently under investigation.

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