Nuclear Alignment and Magnetic Moments of ^{133}Xe, ^{133}Xe^m, and ^{131}Xe^m by Spin Exchange with Optically Pumped ^{87}Rb

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This paper reports the first demonstration of nuclear orientation of radioactive xenon atoms by spin exchange with polarized rubidium atoms. The nuclear tensor alignment, as measured by gamma-ray anisotropy, is about 30% of the maximum possible. The magnetic dipole moments for ^{133}Xe, ^{133}Xe^m, and ^{131}Xe^m have also been measured by NMR spectroscopy. The methods developed here have possibilities for wide applications to nuclear and atomic studies of other radioactive rare-gas atoms.

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In this Letter we describe a new experimental method which has allowed us to produce for the first time large quantities of highly polarized nuclei of radioactive noble gases without the use of low temperatures, high magnetic fields, or Stern-Gerlach magnets. These nuclei have very long spin relaxation times, typically several minutes or longer, and it is therefore possible to obtain magnetic resonance linewidths of a few millihertz, and to make very precise measurements of the magnetic moments of these nuclei and of other small interactions of the nuclei with their environment.

In our work we polarize the nuclei of Xe by spin exchange with Rb atoms which have been electronically spin polarized by optical pumping with a dye laser. The overall spin reaction is

$$\mathrm{Rb}(\uparrow)+\mathrm{Xe}(\downarrow)\rightarrow \mathrm{Rb}(\downarrow)+\mathrm{Xe}(\uparrow), \quad (1)$$

where the arrows in parentheses represent spin directions.

The polarization of helium atoms by spin exchange with polarized Rb atoms was first observed by Bouchiat, Carver, and Varnum.[1] Grover[2] later demonstrated that the heavy-rare-gas atoms have much larger spin-exchange cross sections and large polarizations for the stable xenon isotopes have been observed. The details of the spin transfer (1) are complicated and most of the spin transfer occurs in RbXe van der Waals molecules.[3] A third body, a N_2 molecule in our experiments, is needed to form the RbXe molecules. The N_2 also quenches the fluorescence of the optically pumped Rb atoms and prevents the reabsorption of fluorescence, which destroys the Rb spin polarization.

The three Xe isotopes were oriented by spin exchange with polarized ^{87}Rb and the resulting nuclear alignment was measured by observing γ-ray anisotropies.[4-6] The angular distribution of γ rays emitted from oriented nuclei can be expressed in a series of even Legendre polynomials $P_k(\cos\theta)$,

$$W(\theta)=1+A_2P_2(\cos\theta)+A_4P_4(\cos\theta)+\ldots, \quad (2)$$

where θ is the angle between the orientation axis and the direction of the γ ray. The coefficient A_k can be expressed as a product, $A_k=\rho_kF_k$, of orientation parameter ρ_k and a function F_k which depends on the spin sequence and multipolarity.[7] The tensor-orientation parameters are defined by

$$\rho_k=(2J+1)^{1/2}\sum_m(-1)^{J-m}\times C(J\,J\,k;m\,-m\,0)a_m, \quad (3)$$

where C is the Clebsch-Gordan coefficient and a_m is the population of the m state, normalized to unity, $\Sigma a_m=1$.

The $\frac{11}{2}^-$ isomers ^{131}Xe^m and ^{133}Xe^m decay to the $\frac{3}{2}^+$ ground states. The leading multipolarity should be $M4$ but $E5$ is also allowed. However, the $E5$ is expected to be small for a single neutron transition ($h_{11/2}\rightarrow d_{3/2}$) and has not been observed in other experiments. We therefore assume a pure $M4$ decay and the series accordingly terminates at $k=8$, with F_0, F_2, F_4, F_6, and F_8 given by 1, -0.8890, $+0.4434$, $+0.0320$, and -0.2624, respectively. A useful feature of this decay is that the γ intensity along the orientation axis is zero if the initial nuclei are all in the $m=+\frac{11}{2}$ state.

The ^{133}Xe nucleus, with spin $\frac{3}{2}^+$, decays by allowed Gamon-Teller β emission to $\frac{5}{2}^+$ levels of ^{133}Cs at 81 keV and 161 keV. These states then decay to the $\frac{7}{2}^+$ ground state by γ emission with $E2/M1$ mixing ratios given by[8] $\delta_{81}=-0.152(2)$ and $\delta_{161}=+0.61(2)$. The initial nuclear orientation of the $\frac{3}{2}^+$ ^{133}Xe is transferred to the $\frac{5}{2}^+$ states by β decay, resulting in alignment $\rho_2(\frac{5}{2}^+)=(\sqrt{14}/5)\rho_2(\frac{3}{2}^+)$ and $\rho_4(\frac{5}{2}^+)=0$. Thus, only the Legendre polynomial P_2 contributes to the angular distribution and with $F_2(81\ \mathrm{keV})=0.06714$ and $F_2(161\ \mathrm{keV})=0.8028$ we have $A_2(81\ \mathrm{keV})=-0.0502\rho_2(\frac{3}{2}^+)$ and $A_2(161\ \mathrm{keV})=+0.601\rho_2(\frac{3}{2}^+)$.

The experimental apparatus is illustrated in Fig. 1. The tunable, Kr^+ laser pumped, dye laser produces

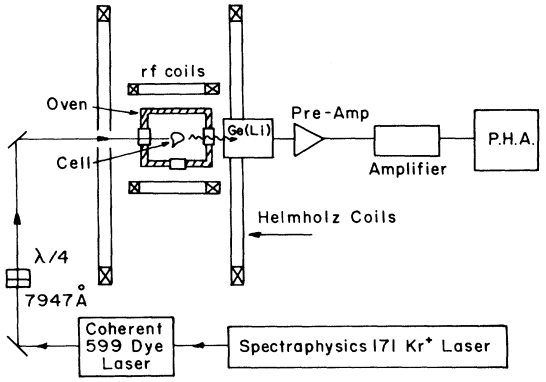

FIG. 1. The experimental apparatus for polarizing Xe isotopes.

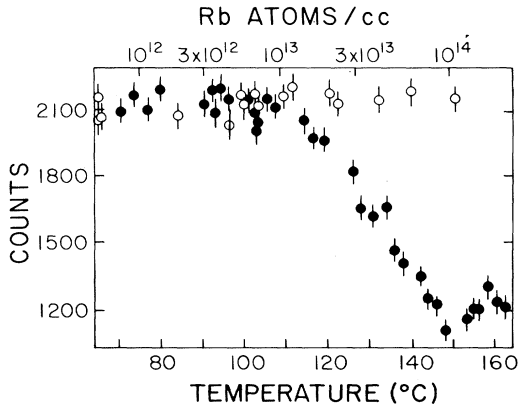

FIG. 2. The count rate from the 164 keV $M4$ γ ray of $^{131}Xe^{m}$ vs cell temperature. The open points were taken with the laser off. The solid points, taken with the laser on, decrease as the Rb density increases, demonstrating significant nuclear orientation.

0.4 W of linearly polarized light at the rubidium D_1 wavelength, 7947 nm, with a bandwidth of ~ 30 GHz. The laser beam is expanded and passed through a $\lambda/4$ plate which converts the light to circular polarization. The second mirror is movable, allowing the cell to be illuminated either with laser light or by a rubidium lamp. The cells, Pyrex-glass spheres with an inner diameter of 1 cm, were filled with a few milligrams of ^{87}Rb metal, about 10 μCi of ^{133}Xe (containing traces of $^{133}Xe^{m}$ and $^{131}Xe^{m}$), and 50 Torr of N_2 gas. The cell was mounted inside an oven heated by flowing hot air. A magnetic field was maintained in the propagation direction of the laser light by three Helmholz coil pairs. A smaller pair of coils near the cell was used to drive magnetic-resonance transitions in the audio- and radio-frequency range.

The γ-ray energy spectrum was measured with a Ge(Li) detector, positioned at $\theta = 0°$, and a multichannel pulse-height analyzer. The intensities in the four peaks at 81 keV, 161 keV, 164 keV, and 233 keV were corrected for background and decay and recorded for each optical pumping and NMR condition. The washout of the angular correlation due to finite detector size is $\sim 5\%$ for the $P_2(\cos\theta)$ term.

Figure 2 illustrates the intensity of the 164 keV line of the $\frac{11}{2}^-$ $^{131}Xe^{m}$ isomer, with laser on and laser off, as a function of cell temperature, or equivalently, Rb density. At room temperature there is no observable difference in count rates indicating no appreciable Xe nuclear alignment. As the temperature is increased the Rb density increases which increases the collision frequency of a Xe atom with polarized Rb atoms. At a temperature of $\sim 110\,°C$ (Rb density of $\sim 10^{13}$ atoms/cm^3), the spin-exchange rate is fast enough to produce noticeable alignment of the xenon nuclei. As the temperature is increased further the γ-ray count rate decreases, as expected with increasing nuclear alignment, reaches a minimum at about 150 °C (10^{14} atoms/cm^3), and then levels off. The maximum fractional change in count rate (ratio of count rate with laser on and laser off) is 0.56, indicating substantial but not complete polarization of the Xe nuclei. The saturation of alignment which occurs at 150 °C is not completely understood at this time but is thought to be due to failure to maintain complete polarization of the Rb atoms at high density (10^{14} atoms/cm^3). At this density, spin destroying Rb-Rb collisions could reduce the Rb spin polarization. To characterize the nuclear orientation we assume that the magnetic substates have an equilibrium Boltzmann distribution given by $P_m \propto e^{\beta m}$ which is determined by a dimensionless spin-temperature parameter β. The maximum alignment achieved ($\sim 150\,°C$) then corresponds to a temperature $\beta = 0.38$. At this temperature the dominant term is the rank-2 alignment for which $\rho_2(\frac{11}{2}^-) \cong 0.51$ while the next term has $\rho_4(\frac{11}{2}^-) \cong 0.04$. The value of $\rho_2(\frac{11}{2}^-)$ if all nuclei were in the $m = +\frac{11}{2}$ state is 1.74. Our result is 29% of this value.

Although we illustrate only the $^{131}Xe^{m}$ data we obtain substantially the same alignment for both $\frac{11}{2}^-$ isomers, $^{131}Xe^{m}$ and $^{133}Xe^{m}$. The spin $\frac{3}{2}^+$ ^{133}Xe, however, behaves a little differently. We find that the alignment begins to be noticeable at a slightly lower temperature, namely $\sim 100\,°C$ instead of 110 °C as obtained for the $\frac{11}{2}^-$ isomers. This is not unexpected since the gyromagnetic ratio of ^{133}Xe is larger than that for the $\frac{11}{2}^-$ isomers (see below) and this should enhance the spin-transfer process. The ^{133}Xe alignment increases with temperature up to 140 °C and then begins decreasing with higher temperature, unlike the $\frac{11}{2}^-$ isomer which seems to level off. At the peak in orientation the rank-2 alignment parameter is $\rho_2(\frac{3}{2}) \approx 0.28$ which corresponds to a spin-temperature parameter of $\beta(\frac{3}{2}) \approx 0.83$. If all ^{133}Xe nuclei were in

the $m = +\frac{3}{2}$ state the value of $\rho_2(\frac{3}{2})$ would be 1.0. We have achieved, therefore, about 30% of this maximum for ^{133}Xe.

The magnetic-dipole moments of the xenon isotopes were measured by NMR spectroscopy. A weak alternating magnetic field B_{rf} was applied tranverse to the static field B_0. The resonances were observed by scanning the frequency of the alternating field under conditions in which the cell was at $\sim 150\,^{\circ}$C and the Xe nuclei were oriented by the laser. When the rf frequency coincides with the nuclear-precession frequency the nuclear orientation is destroyed and the count rate changes. The resonance pattern for ^{131}Xe^m is illustrated in Fig. 3. We note that the 0.5-Hz width of the resonance is determined in this case by the strength of the rf field but much narrower resonances are possible at lower rf fields because of very long spin-relaxation times. To minimize the effects of long-term drifts in the magnetic field, the data were taken in three sets of resonance pairs, namely, ^{133}Xe-^{131}Xe^m, ^{133}Xe^m-^{131}Xe^m, and ^{87}Rb-^{131}Xe^m. The resonance data for each pair were also interwoven so that the ratio of the two resonance frequencies would be accurate. The resonance frequencies are given in Table I.

The frequencies for Zeeman transitions of ^{87}Rb, which were used for field calibration, were measured by observing changes in the light transmitted through the cell when the rf field induced Zeeman transitions in the $F=2$ multiplet. A rubidium lamp was used for these measurements because the laser light is too noisy. Since the low light intensity of the lamp is unable to penetrate the dense Rb vapor at $\sim 150\,^{\circ}$C the cell had to be quickly cooled to $\sim 90\,^{\circ}$C for the measurements.

The ^{87}Rb Zeeman transitions are not resolved at the field used (≈ 2 G). The observed resonance line is thus a superposition of all four transitions in the multiplet $F=2$. The line has an asymmetric shape which reverses with the circular polarization of the light. An average over all four transitions and both polarization states yields the frequency given in Table I. This corresponds to a field experienced by the Rb atoms of $B_0 = 2.151\,72(5)$ G. There is an additional small contribution to the field experienced by the xenon atoms due to collisions with polarized Rb atoms. From independent studies[9] we estimate that this field would be $B_{\text{Rb-Xe}} = 3.09(30)\times 10^{-3}$ G with 100% polarization at the Rb density corresponding to a cell temperature of $147 \pm 5\,^{\circ}$C. As a result of the uncertainty in the Rb polarization at high density, we take the average field to be 75% ±25% of this maximum value. (Collisions of Xe with other Xe atoms and with N_2 molecules can be expected to cause a frequency shift of 10^{-4} Hz or less, based on the work of Brinkman *et al.*[10]) The total field experienced by the Rb atoms in run 3 is therefore $B_{\text{Xe}} = 2.154\,04(80)$ G. This value was used to determine the magnetic moment of ^{131}Xe^m from the data of run 3. Runs 1 and 2 then determine the magnetic moments of ^{133}Xe and ^{133}Xe^m. Table II summarizes the magnetic-moment values with and without the correction for atomic diamagnetism. We have not determined the signs of the moments. Previous results are also given in Table II. The present results are substantially more accurate for the $\frac{11}{2}^-$ isomers than the previous values which were obtained by low-temperature methods. Our ^{133}Xe result agrees reasonably well with a preliminary value obtained by laser hyperfine studies in excited Xe states.

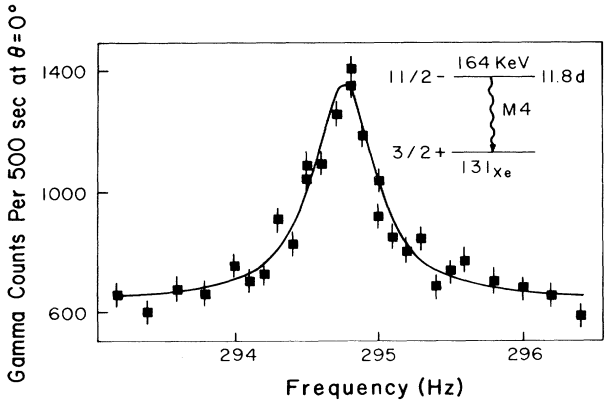

FIG. 3. The NMR pattern for the $\frac{11}{2}^-$ isomer ^{131}Xe^m. The 0.5-Hz width of the resonance is due to power broadening.

TABLE I. Centroid frequencies for spin resonances.

Run	Xe	ν_x (Hz)	$\nu(^{131}\text{Xe}^m)$ (Hz)	$\nu_x/\nu(^{131}\text{Xe}^m)$
1	^{133}Xe	884.442(77)	294.781(6)	3.000 34(27)
2	^{133}Xe^m	321.077(45)	294.850(8)	1.088 95(16)
3	^{87}Rb	1505 291(32)	294.753(8)	5106.96(18)

TABLE II. Magnetic moments.

Isotope	$\lvert\mu_{\text{uncorr}}\rvert$ (μ_N)	$\lvert\mu_{\text{corr}}\rvert$ [a] (μ_N)	μ_{previous}
^{131}Xe^m	0.987 34(38)	0.994 35(38)	−0.80(10) [b]
^{133}Xe	0.807 92(32)	0.813 65(32)	0.8125(3) [c]
^{133}Xe^m	1.075 16(45)	1.082 79(45)	−0.87(12) [b]

[a]Corrected for diamagnetism with $1/(1-\sigma) = 1.007\,092$. See Ref. 11.

[b]Reference 12.

[c]Reference 13.

These methods can be extended to other radioactive Xe isotopes and very likely to radioactive isotopes of Kr and Rn. The already high spin polarization can certainly be increased by more careful attention to wall relaxation and to the basic limits of optical pumping at high alkali-vapor densities. The radioactive isotopes have already been useful for studying spin polarization in samples which are too optically thick for conventional optical methods.

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