NUCLEAR MOMENT ALIGNMENT, RELAXATION
AND DETECTION MECHANISMS

February 1980 - January 1981
ANNUAL TECHNICAL REPORT

Contract No. F49620-77-C-0047

Prepared for
AIR FORCE OFFICE OF SCIENTIFIC RESEARCH

GUIDANCE & CONTROL SYSTEMS
LH-4, 8900 Canoga Avenue, Woodland Hills, California 91365

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**Title:** Nuclear Moment Alignment, Relaxation and Detection Mechanisms

**Authors:** Tae M. Kwon

**Abstract:**

The reported physics research is part of an overall program to develop a nuclear magnetic resonance gyro that makes use of an optically pumped alkali metal vapor both to align the magnetic moments of the noble gas nuclei and to detect the weak magnetic fields that are generated by these precessing nuclear moments.

Theoretical and experimental investigations are reported on the relaxation of polarized nuclear spins of Xe$^{131}$ through the interaction of the nuclear quadrupole moment with the electric field gradient. The field gradient arises from deformation of the otherwise spherical electronic shell of the xenon...
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Approved By:

Emery L. Moore
Manager, NMR Gyro Project

GUIDANCE & CONTROL SYSTEMS
Litton 5500 Canoga Avenue, Woodland Hills, California 91365
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SECTION I
PROGRAM DESCRIPTION

1.1 INTRODUCTION

The Guidance and Control Systems Division of Litton Systems, Inc. is involved in a multi-year program in the research and development of a nuclear magnetic resonance (NMR) gyro for use in inertial navigation systems.

In the magnetic resonance gyro, angular craft rate is sensed through a shift in the precession frequency of an ensemble of atomic or nuclear magnetic moments. The Litton NMR gyro utilizes ensembles of two noble gas isotopes in order to eliminate explicit dependence on the magnetic field. In order to obtain the gyroscopic information in this scheme, the noble gas ensembles must first be polarized so that coherent precession about an applied magnetic field can take place and, secondly, a means for detecting the nuclear precession must be provided. In the Litton approach, the noble gas ensembles attain a net magnetic moment through spin exchange with an optically oriented alkali vapor. In addition, the alkali vapor, through a magnetometer effect, detects the weak magnetic fields, which are generated by the precessing nuclear moments of the two noble gases, and thus provides the gyro angular rate information.

The purpose of the physics research program is to develop a stronger theoretical and empirical understanding of the alkali-atom noble-gas-atom system. Of particular interest are the spin exchange interactions between the alkali valence electrons and the noble gas nuclei, the noble gas nuclear spin relaxation mechanisms as they pertain to the NMR gyro and other related phenomena which impact the NMR gyro technology.
1.2 OBJECTIVES AND STATUS OF THE CURRENT PROGRAM

The research tasks described in Sections II and III of this report are part of our continuing effort to understand and parameterize the alkali-atom noble-gas-nucleus spin exchange interaction. In addition, investigations of the spin exchange process also provide opportunities to study the pertinent relaxation mechanisms of the noble gas nuclei.

Reported in Section II are the results of the theoretical and experimental investigation on the relaxation of polarized noble gas nuclei through the interaction of the nuclear quadrupole moment with the electric field gradient arising from collisions with the walls. In the last AFWAR annual Technical Report, a simple theoretical model of this effect was developed and was in agreement with the experimental data only qualitatively. No quantitative analysis of the data was possible with the model. In the present work, the model has been extended to incorporate the second order expansion of the nuclear density matrix. Under this new model, measurements of the relaxation of Xe\(^{131}\) nuclei were analyzed quantitatively, and, as a result, the sticking time of the Xe\(^{131}\) atom onto the container wall and the strength of the nuclear quadrupole interaction have been estimated for the first time.

In Section III, we report on our measurement of the shift in the Larmor precession frequency of the noble gas nuclei during spin exchange with an optically oriented alkali vapor. The results confirm that, for a given alkali polarization environment, the difference in frequency shifts for different isotopes of the same noble gas atom, such as Xe\(^{129}\) and Xe\(^{131}\), is smaller by, at least, an order of magnitude than the difference in frequency
shifts for different noble gas species, such as $^1_{29}\text{Xe}$ and $^8_{3}\text{Kr}$. Experimental results are presented in terms of the parameters relevant to the NMR gyro.

Investigations into this area of physics have relevance not only to NMR gyro research but also to other possible areas of research such as magnetometers, atomic clocks, masers, laser stabilization, and polarized nuclear targets. It was in this light that the result of the work reported in Section II has been prepared as a scientific paper to be submitted for publication to The Physical Review. The preprint of this paper is included as paragraph 2.2.

In the course of this work, an idea was conceived to coat the container wall with RbH to reduce the nuclear quadrupole interaction. Successful implementation of the idea has been demonstrated in the data reported in Section II. We believe that the invention of the coating material and coating technique is a significant step forward in the development of NMR devices. It is for this reason that we have submitted a written disclosure of invention to the company for consideration in applying for a patent. The patent application is under consideration for further action by the corporate headquarters.

1.3 PROFESSIONAL PERSONNEL ASSOCIATED WITH THE PROGRAM

The principal investigator of these research efforts has been Dr. Tae M. Kwon. This work has also been strongly supported by Dr. Charles H. Volk, and Dr. John G. Mark. We also wish to acknowledge the contribution of Mr. Howard E. Williams for the design and Mr. William P. Debley for the construction of various components of the apparatus, including the resonance cells and lamps used in this research effort. Finally we acknowledge Dr. Edward Kanegsberg for many helpful discussions.
SECTION II
QUADRUPOLAR NUCLEAR SPIN RELAXATION

2.1 INTRODUCTION

Two nuclear species, Xe$^{129}$ (I=1/2) and Xe$^{131}$ (I=3/2), or Xe$^{129}$ and Kr$^{83}$ (I=9/2) are required under Litton's approach to the development of a nuclear magnetic resonance (NMR) gyro. Associated with the nuclei of Kr$^{83}$ and Xe$^{131}$, and characteristic of nuclear spins greater than 1/2, is the nuclear quadrupole moment which interacts with electric field gradients. Field gradients arise at the nuclear site during collisions of noble gas atoms with the walls of their containers. This interaction is more pronounced in the Xe$^{131}$ nuclear ensemble than in Kr$^{83}$. Attention is directed to the fact that, limited by this interaction, the transverse relaxation time ($T_2$) of the Xe$^{131}$ nuclei published in available literature prior to this contract had been on the order of a few hundred milliseconds.* At the initial stage of this contract, it was improved to 5 to 10 seconds in 15 ml spherical samples. The extremely short $T_2$ made it difficult to improve our understanding on the subject of the alkali-atom-noble-gas-nucleus spin exchange interaction, which is essential for the development of an NMR gyro.

Both theoretical and experimental understanding of the nuclear quadrupole interaction continues to improve. In this section, we report $T_2$ of a Xe$^{131}$ nuclear ensemble as long as 220 seconds. This improvement was accomplished by coating the cell wall with RbH. With the experimental advantage of a larger $T_2$, the parameters relevant to the nuclear quadrupole interaction were estimated for the first time. The results of investigations

have been prepared as a scientific paper for consideration of publication in the Physical Review. The preprint of this paper is included in the Appendix to this section in lieu of a paraphrased report on the research performed under this contract. The developed process of data analysis can now be utilized routinely in the gyro development to determine the characteristics of cells containing $^{131}$Xe$^{113}$ (or $^{83}$Kr$^{83}$) nuclei.

Investigations of noble gas nuclear quadrupole moments interacting with the walls of their container will continue, particularly for RbH coated glass walls. In order to improve the uniformity of the coating, the reaction will be studied in glass containers made of other than pyrex glass. Non-pyrex glass will be studied because the rubidium reaction with hydrogen molecules has been shown to be sensitive to the presence of impurities (such as water, oxygen, etc.) normally embedded on pyrex glass.*

2.2 PUBLICATION PREPRINT

A copy of a preprint to be submitted to the Physical Review for consideration of publication follows herein.

Quadrupole nuclear spin relaxation of $^{131}\text{Xe}$ in the presence of rubidium vapor

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(Received

Nuclear spin relaxation of $^{131}\text{Xe}$ produced by collisions of the xenon atoms with the walls of the experimental cell has been studied. The xenon nuclear ensemble is polarized through spin exchange collisions with an optically oriented rubidium vapor. Depolarization of the $^{131}\text{Xe}$ ensemble in the wall collisions is assumed to be due to the quadrupole coupling between the $^{131}\text{Xe}$ nucleus and the electric field gradients which appear at the nucleus during collisions of the atoms with the cell walls. Model equations describing the nuclear relaxation are derived and estimates of the electric field gradients are given.

PACS numbers: 76.60.Gv, 76.60.Es, 07.58.+g
I. INTRODUCTION

We recently reported on measurements of the transverse nuclear spin relaxation of $^{83}\text{Kr}$ in the presence of rubidium vapor. The decay of the krypton nuclear spin was found to be dependent on the orientation of the experimental cell with respect to an axis defined by the static magnetic field present in the apparatus. The angle between the cell and the magnetic field which yielded the longest decay time was termed the 'magic' angle after similar work done on $^{201}\text{Hg}$\(^2\). Furthermore, we observed that the decay signal, off the magic angle, possessed a non-exponential character. These effects were modeled as an interaction between the nuclear quadrupole moment and the electric field gradients present at the krypton nucleus during collisions of the krypton atoms with the walls of the experimental cell. The model describes the transverse nuclear relaxation by $2I$ precessional frequencies, where $I$ is the nuclear spin; $I = \frac{9}{2}$, for $^{83}\text{Kr}$. We are able to show a qualitative agreement between this model and the experimental measurements. A quantitative analysis of the krypton data was not possible due to the complexity of the signal.

We report here experimental data on the transverse nuclear spin decay of $^{131}\text{Xe}$ (nuclear spin, $I = \frac{3}{2}$), which exhibits relaxation through an electric quadrupolar interaction in collisions with the cell walls. In order to simplify the experimental data analysis we have re-formulated our model of the nuclear quadrupolar interaction, viewing the precession of the nuclear ensemble as $2I$ oscillators each with a single frequency. In this way we treat $2I$ two-state Bloch type systems\(^3\), instead of treating one complex system of $2I$ frequencies. This procedure is similar to the 'ficticious spin-1/2
operators' presented by Vega and Pines. Furthermore, our previous model is extended in this work by incorporating results of a second order expansion of the nuclear density matrix as given by Cohen-Tannoudji for the case of relaxation of nuclear spin 3/2 under a quadrupolar interaction. In this new model, we show that the second order expansion leads to a coupling among the 2I oscillators.

II. THEORETICAL MODEL

We consider in this section the transverse relaxation of a nuclear spin ensemble of nuclear spin $I = 3/2$ under a quadrupolar interaction. We will rely on results presented in Refs. (1 and 5) and repeat only selected details here, as clarity demands.

A. The energy eigenvalues. The full spin Hamiltonian of the system is taken to be:

$$H = H_M + H_Q$$

where $H_M$ is the static magnetic interaction

$$H_M = -\hbar H_o I_z$$

with the nuclear gyromagnetic ratio, $H_o$ the external magnetic field, taken to be along the z-axis, and $I_z$, the z-component of nuclear spin. $H_Q$, the quadrupolar Hamiltonian, represents the interaction of the nuclear electric quadrupole moment with an electric field gradient. As was argued in Ref. (1), the electric field gradient arises during collisions of the noble gas atoms with the walls of the experimental cell. If we assume that for a given collision, the resultant electric field
gradient possesses cylindrical symmetry, then the quadrupole Hamiltonian can be written as:\textsuperscript{1,5,6}

\[
H_Q = \frac{e^2Qq}{4\mathrm{i}(2I-1)} \left\{ \frac{1}{2} (3 \cos^2 \theta - 1) \left[ 3I_z^2 - I(I+1) \right] \right.
\]

\[
+ \frac{3}{2} \sin \theta \cos \theta \left[ I_z (I^+ + I^-) + (I^+ I^-) I_z \right] + \frac{3}{4} \sin^2 \theta \left( I^+ I^- + I^- I^+ \right) \}
\]

(3)

where \( e \) is the electronic charge, \( e^2V/\partial z^2 \), is the electric field gradient under the assumption of cylindrical symmetry, \( Q \) is the quadrupole moment of the noble gas nucleus, \( \theta \) is the angle between the quantization axis and the cylindrical axis of the electric field gradient, and \( I^\pm \) are the raising and lowering operators associated with the spin operator, \( I \), where in the cartesian reference:\textsuperscript{7}

\[
I^\pm = I_x \pm \mathrm{i} I_y .
\]

(4)

As in Ref. (1), we make the following definitions:

\[
a = (3\cos^2 \theta - 1), \quad (5a)
\]

\[
b = \sin \theta \cos \theta, \quad (5b)
\]

\[
c = \sin^2 \theta, \quad (5c)
\]

\[
a = e^2Qq/4\mathrm{i}(2I-1), \quad (5d)
\]

\[a = e^2Qq/4\mathrm{i}(2I-1), \quad (5d)
\]

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\]
The quadrupole Hamiltonian, $H_Q$, is small compared to the magnetic Hamiltonian $H_M$, and thus we compute the energy eigenvalues for the full Hamiltonian employing perturbation theory. The energy levels for a nucleus with spin 3/2 are computed through second order:

$$E_1 = \frac{3}{2} \omega + \frac{3}{2} \alpha a - \frac{27 \alpha^2 b^2}{\omega} + \frac{27 \alpha^2 c^2}{8\omega}$$

(6a)

$$E_2 = \frac{1}{2} \omega - \frac{3}{2} \alpha a - \frac{27 \alpha^2 b^2}{\omega} + \frac{27 \alpha^2 c^2}{8\omega}$$

(6b)

$$E_3 = -\frac{1}{2} \omega - \frac{3}{2} \alpha a + \frac{27 \alpha^2 b^2}{\omega} - \frac{27 \alpha^2 c^2}{8\omega}$$

(6c)

$$E_4 = -\frac{3}{2} \omega + \frac{3}{2} \alpha a - \frac{27 \alpha^2 b^2}{\omega} - \frac{27 \alpha^2 c^2}{8\omega}$$

(6d)

B. The system equations. The evolution of the observables of the system are found by solving the density matrix equation of the ensemble, which is given by:

$$\frac{d\rho}{dt} = \frac{i}{\hbar} [\rho, H]$$

(7)

and then computing the expectation value for a system observable, as:

$$\langle O \rangle = \text{Tr} [\rho, O]$$

(8)
where $H$ is the Hamiltonian of the system given in Eq. (1), $\rho$ is the density matrix of the nuclear ensemble and $O$ is an operator corresponding to an observable of the system. For the situation represented by Eq. (1), in which there is a large time-independent interaction, $H_M$, and a much smaller but time-dependent term, $H_Q(t)$, it is convenient to transform the density matrix into the interaction representation, which is defined by:

$$p(t) = e^{-(i/\hbar)H_M t} \rho^*(t) e^{(i/\hbar)H_M t}$$  \(9\)

where $\rho^*(t)$ represents the density matrix in the interaction representation. The differential equation for $\rho^*$ is easily found to be:

$$\frac{d\rho^*}{dt} = \frac{i}{\hbar} [\rho^*(t), H^*_Q(t)]$$  \(10\)

where $H^*_Q$ is given by:

$$H^*_Q(t) = e^{-(i/\hbar)H_M t} H_Q(t) e^{(i/\hbar)H_M t}$$  \(11\)

The time dependence of $H_Q$ is taken to be that due to stochastic collisions of the xenon atoms with the walls of the experimental cell, in the same manner as that treated in Ref. (5).
In the limit that the quadrupole interaction can be taken as a perturbation, we solve Eq. (10) in an iterative manner, which through second order is given by:

\[
\frac{d\rho}{dt} = \frac{i}{\hbar} \left[ \rho^{*}(0), H_{Q}^{*}(t) \right] + \left( \frac{i}{\hbar} \right)^{2} \int_{0}^{t} \left\{ \left[ \rho^{*}(0), H_{Q}^{*}(t') \right], H_{Q}^{*}(t) \right\} dt'. \tag{12}
\]

The first term in Eq. (12) was shown previously to lead to a transverse decay of the nuclear ensemble through a dephasing of the precessing Zeeman levels. The second term in Eq. (12) has been treated by Cohen-Tannoudji for the case of relaxation of \(^{201}\text{Hg}\) under a quadrupole interaction. Since the nuclear spin of \(^{201}\text{Hg}\) is 3/2, we can directly apply the results of Cohen-Tannoudji's work to our analysis of \(^{131}\text{Xe}\).

The commutators in Eq. (12) are evaluated under the following assumptions:

1. The first order commutator can be evaluated with a diagonal Hamiltonian whose eigenvalues are computed through second order; and,
2. The square of the product of the correlation time of the interaction, and the energy level separations of the nuclear Zeeman states, is much less than unity.

The first term in Eq. (12) is then evaluated by employing some straightforward matrix algebra, while the evaluation of the
second terms is found in Ref. (5). Eq. (12) is then written, transformed back into the Schroedinger representation, as:

\[
\begin{bmatrix}
    0 & -\Delta W_{12}\rho_{12} & -\Delta W_{13}\rho_{13} & -\Delta W_{14}\rho_{14} \\
    \Delta W_{12}\rho_{21} & 0 & -\Delta W_{23}\rho_{23} & -\Delta W_{24}\rho_{24} \\
    \Delta W_{13}\rho_{31} & \Delta W_{23}\rho_{32} & 0 & -\Delta W_{34}\rho_{34} \\
    \Delta W_{14}\rho_{41} & \Delta W_{24}\rho_{42} & \Delta W_{34}\rho_{43} & 0
\end{bmatrix}
\]

(13)

where the first matrix in Eq. (13) is the result of the evaluation of the first order expansion in Eq. (12), with \(\Delta W_{ij} = (E_i - E_j)/\hbar\), where the bar represents a time average over the \(ij\) collision. The second matrix in Eq. (13) is taken from the results in Ref. (5), where from assumption 2 above, we have:

\[
1/T_Q = J_0 = J_1 = J_2.
\]

(14)
The $J_i$'s are the notation in Ref. (5) for the general decay rates due to a quadrupolar interaction. The condition in Eq. (14) results in a single exponential decay for the nuclear ensemble when the cell is oriented at the magic angle. This is observed experimentally.

The observable of interest in this particular experiment is the $x$-component of the noble-gas polarization $^{1,12}$, $<I_x>$. The corresponding operator for the spin 3/2 system, written in matrix form is:

$$
I_x = (1/2) \begin{bmatrix}
0 & \sqrt{3} & 0 & 0 \\
\sqrt{3} & 0 & 2 & 0 \\
0 & 2 & 0 & \sqrt{3} \\
0 & 0 & \sqrt{3} & 0
\end{bmatrix}
$$

(15)

The expectation value, $<I_x>$, is given by:

$$
<I_x> = Tr \left[ I_x \rho \right]
$$

(16)

For the spin 3/2 system, Eq. (16) becomes:

$$
<I_x> = \frac{\sqrt{3}}{2} (\rho_{12} + \rho_{21}) + 2(\rho_{23} + \rho_{32}) + \frac{\sqrt{3}}{2} (\rho_{43} + \rho_{34})
$$

(17)
where the \( p_{ij} \)'s are the elements of the density matrix, with 
\( p_{ii} \) the population density of the state whose energy eigenvalue 
is given by \( E_i \) in Eq. (6). With the following definition:

\[
M_{x1} = \frac{3}{2} (p_{12} + p_{21}) \quad (18a)
\]

\[
M_{x2} = (p_{23} + p_{32}) \quad (18b)
\]

and,

\[
M_{x3} = \frac{3}{2} (p_{34} + p_{43}) \quad (18c)
\]

we can write Eq. (17) as:

\[
\langle I_x \rangle = M_{x1} + M_{x2} + M_{x3} \quad (19)
\]

We have adopted the symbol 'M' for the sake of similarity to the
Bloch equations. The M's here represent a two-state polarization
of the four state spin 3/2 system. In exactly the same
manner, we can express the y-component of noble-gas polarization
in terms of the two state polarizations, \( M_{y1}', M_{y2} \) and \( M_{y3}' \)
appropriately defined. Using Eqs. (13 and 17-19) it is only a
matter of algebra to show that the following set of equations
describe the evolution of the nuclear ensemble under the
quadrupolar interaction in terms of the two state polarizations:

\[
\begin{bmatrix}
\dot{M}_{x1} \\
\dot{M}_{y1} \\
\dot{M}_{x2} \\
\dot{M}_{y2} \\
\dot{M}_{x3} \\
\dot{M}_{y3}
\end{bmatrix} =
\begin{bmatrix}
-3/T_Q & -\Delta W_{12} & 0 & 0 & 1/T_Q & 0 \\
\Delta W_{12} & -3/T_Q & 0 & 0 & 0 & 1/T_Q \\
0 & 0 & -2/T_Q & -\Delta W_{23} & 0 & 0 \\
0 & 0 & \Delta W_{23} & -2/T_Q & 0 & 0 \\
1/T_Q & 0 & 0 & 0 & -3/T_Q & -\Delta W_{34} \\
0 & 1/T_Q & 0 & 0 & \Delta W_{34} & -3/T_Q
\end{bmatrix}
\begin{bmatrix}
M_{x1} \\
M_{y1} \\
M_{x2} \\
M_{y2} \\
M_{x3} \\
M_{y3}
\end{bmatrix}
\]

(20)
Eq. (20) describes the evolution of a set of three oscillators. The coupling, which results from the inclusion of the second order terms, is between the first and third oscillators. If we neglect the coupling terms, the system equations are easily seen to reduce to the case treated in Ref. (1), with the solution given by Eq. (18) of Ref. (1). In addition, the uncoupled equations can each be written in the form:

\[ M = (W \times M) + M/T_Q \]  

which is exactly the form of the Bloch equations.²

Due to other relaxation effects on the $^{131}$Xe ensemble, we have added an additional phenomenological term to all the diagonal elements. This procedure agrees with the results we have seen for the situation in which the quadrupole interaction is negligibly small.¹³ The system of equations for the two-state polarizations then become:

\[
\begin{bmatrix}
\dot{M}_{x1} \\
\dot{M}_{y1} \\
\dot{M}_{x2} \\
\dot{M}_{y2} \\
\dot{M}_{x3} \\
\dot{M}_{y3}
\end{bmatrix} = 
\begin{bmatrix}
\frac{3}{T_Q} - \frac{1}{T_2} & -\Delta W_{12} & 0 & 0 & \frac{1}{T_Q} & 0 \\
\Delta W_{12} - \frac{3}{T_Q} - \frac{1}{T_2} & 0 & 0 & 0 & \frac{1}{T_Q} & 0 \\
0 & 0 & -\frac{2}{T_Q} - \frac{1}{T_2} & -\Delta W_{23} & 0 & 0 \\
0 & 0 & \Delta W_{23} - \frac{2}{T_Q} - \frac{1}{T_2} & 0 & 0 & 0 \\
\frac{1}{T_Q} & 0 & 0 & 0 & -\frac{3}{T_Q} - \frac{1}{T_2} & -\Delta W_{34} \\
0 & \frac{1}{T_Q} & 0 & 0 & \Delta W_{34} - \frac{3}{T_Q} - \frac{1}{T_2} & 0
\end{bmatrix}
\begin{bmatrix}
M_{x1} \\
M_{y1} \\
M_{x2} \\
M_{y2} \\
M_{x3} \\
M_{y3}
\end{bmatrix}
\]  

(22)
where $1/T_2$ is the relaxation rate due to the other processes present in the cell, for instance, spin exchange of the $^{131}\text{Xe}$ nuclear spin with the rubidium valence electron. 13

III. EXPERIMENT

The experimental apparatus and the generation of the signal from the noble gas nuclear ensemble has been previously discussed in detail. 14 We will describe here some of the salient features of the experimental set-up for the sake of the continuity of this report.

The experimental apparatus is shown schematically in Figure 1. The heart of the apparatus is the experimental cell, which is a 1-ml Pyrex sphere. The cell is sealed on a vacuum system with an excess of natural rubidium metal, 0.1 Torr enriched $^{129}\text{Xe}$, 0.4 Torr enriched $^{131}\text{Xe}$, 10 Torr $\text{N}_2$, 100 Torr $\text{He}$ and 10 Torr $\text{H}_2$. Enriched $^{129}\text{Xe}$ consists of 61.22 mole % of $^{129}\text{Xe}$ and 13.44 mole % $^{131}\text{Xe}$. Enriched $^{131}\text{Xe}$ contains 63.89 mole % $^{131}\text{Xe}$ and 0.7 mole % $^{129}\text{Xe}$. Remaining components of each of the enriched species consist of even isotopes of xenon. 15 Since there is a $^{129}\text{Xe}$ impurity in enriched $^{131}\text{Xe}$, enriched $^{129}\text{Xe}$ is added to the cell to ensure a sufficient $^{129}\text{Xe}$ signal so that the effects of the $^{129}\text{Xe}$ on the signal can be properly removed in the data analysis. The $\text{N}_2$ and the $\text{He}$ are present to maximize the rubidium polarization, which is used to polarize the Xe noble gas ensembles through spin exchange. 14

In order to simplify analysis of the experimental data in the light of the theory developed in Section II, the electric field gradient experienced by the ensemble of $^{131}\text{Xe}$ nuclei during collisions with the cell wall must be nearly cylindrically symmetric. It was found in the course of this work, that a cell
exhibiting field gradients of a cylindrically symmetric nature could be made by adding $H_2$ to the cell and baking the cell at $80^\circ$C for about two weeks. During the baking period, the rubidium metal was held in the small region of the tip-off area of the spherical cell by forcing the tip region to be cool relative to the cell walls. Pyrex walls of the cells so processed were covered with milky particles, which we later identified as RbH. Excess rubidium metal was still present in the cell. When the cell wall was coated uniformly with RbH, the imaginary axis diametrically through the tip of the cell was found to correspond to the axis of cylindrical symmetry of the quadrupolar interaction. This condition was verified experimentally if a cell showed the magic angle behavior when the angle between the magnetic field and the imaginary axis through the tip was approximately $55^\circ$. No such symmetry could be observed in the cells for which no processing was done. It was further noted that $^{131}$Xe polarization is much higher in the RbH coated cells than in the cells without coating.

Within the experimental apparatus the cell is centered in a resistance-heated oven, provided with Pyrex windows at either end. The oven controls the temperature to within $\pm0.1^\circ$C with a temperature uniformity across the cell of about $0.5^\circ$C. The cell is mounted in a holder which allows it to be rotated in two dimensions from outside the apparatus with a precision of 0.5 deg. The oven is contained in a cylindrical coil form, providing three mutually perpendicular Helmholtz coils. The oven assembly is within four cylindrically concentric magnetic shields. These shields reduce the external magnetic fields to below 10 $\mu$G.

The instantaneous transverse polarization of the noble gas ensembles are determined by measuring the effective magnetic fields of these polarized ensembles. The nuclear ensembles are
polarized in spin exchange collisions with the optically oriented rubidium vapor. The rubidium vapor is optically pumped by illuminating the cell with D₁ circularly polarized light from a rubidium discharge lamp. Detection of the effective magnetic fields generated by the polarized nuclear ensembles is accomplished by utilizing the rubidium vapor as a magnetometer. This technique has been thoroughly discussed in the literature \(^{18,19}\) and our particular mechanization has been previously described.\(^{14}\) The axes for this work have been labelled in a slightly different manner than what was done in Ref. (14). This change was made to facilitate the form of the model equations and presents no difficulty in applying the results as displayed in Ref. (14) to this work.

The particular experimental procedures for this work are as follows. In the presence of the light, a 2 mG field is applied initially along the y-axis, as defined in Figure 1. This condition continues for 10 min., in which time a significant noble gas nuclear polarization is built up. The y-field is then switched to zero while a precessional magnetic field of 350 \(\mu\)G is applied along the z-axis together with an ac magnetic field \((H_{ac})\) along the x-axis, which defines the sensitive axis of the rubidium magnetometer. The cell is arranged in the apparatus with the cell's imaginary axis in the x-z plane. The imaginary axis of the cell can be rotated a full 360 degrees in this plane. As we noted earlier, the imaginary axis diametrically through the tip of the cell has been found to be the cylindrically symmetric axis of the quadrupole interaction. Rotation of the imaginary axis in the x-z plane then corresponds to a rotation of the cell through the angle as defined in Eq. (3). There is an additional ability to rotate the cell's imaginary axis \(\pm 5\) degrees out of the x-z plane. This feature was employed to check the assumption of cylindrical symmetry of the interaction. The
geometry of the cell is shown in Figure 2. 4000 data points at equal time intervals over 300 seconds are taken after the y-magnetic field is switched off. Angle data at 10 deg. cell angle increments in the x-z plane was taken. The data from the first half cycle, 0 to 180 deg. is presented in this report. All data in this report were acquired at a cell temperature of 81.3°C ±0.5°C.

IV. ANALYSIS

A. The signal form. In order to facilitate the data reduction, we re-write the form of the two-state polarization equations from that given in Section II. For brevity, we introduce the following notation:

\[ \omega_c = \omega + \frac{54b^2a^2}{\omega} \quad (23a) \]
\[ \omega_1 = \Delta W_{12} = \omega_c + 3\bar{a} \quad (23b) \]
\[ \omega_2 = \Delta W_{23} \quad (23c) \]
\[ \omega_3 = \Delta W_{34} = \omega_c - 3\bar{a} \quad (23d) \]

where \( \bar{a} \) represents a time average of \( a \) over the collisions. Eq. (22) can be conveniently broken in two parts, due to the particular coupling among the oscillators. One equation to represent the second oscillator, subscripted '2', with frequency \( \omega_2 \) and a set of equations for oscillators '1' and '3', with frequencies \( \omega_1 \) and \( \omega_3 \), respectively. We take advantage of the symmetric separation in these two oscillator frequencies from
the center frequency, $\omega_c$, and write $M_x$ for oscillator 1 and 3 as:

$$M_{xs} = M_x^0 e^{-(p+r)t} F(\theta) \sin(\omega_c t)$$  \hspace{1cm} (24)$$

$$F(\theta) = \begin{cases} 
\cosh(\epsilon t) + (r/\epsilon) \sinh(\epsilon t) & r > 3\bar{a} \\
1 + rt & r = 3\bar{a} \\
\cos(\epsilon t) + (r/\epsilon) \sin(\epsilon t) & r < 3\bar{a} 
\end{cases}$$  \hspace{1cm} (25a)$$

$$p = \frac{1}{T_2} + \frac{2}{T_Q}$$  \hspace{1cm} (25b)$$

$$r = \frac{1}{T_Q}$$  \hspace{1cm} (25c)$$

$$\epsilon^2 = |r^2 - (3\bar{a})^2|$$  \hspace{1cm} (25d)$$

where the subscript 's' denotes either 1 or 3 and $M_x^0$ is the initial amplitude of the signal for these oscillators. The frequency of the second oscillator, $\omega_2$, can be written as:

$$\omega_2 = \omega_c + \Delta \omega_2$$  \hspace{1cm} (26)$$

where

$$\Delta \omega_2 = -\frac{108\bar{a}^2 b^2}{\omega} + \frac{27\bar{a}^2 c^2}{4\omega}.$$  \hspace{1cm} (27)$$
404577

\( \Delta \omega_2 \) is due to a second order effect, which is negligibly small. In this limit we can write the expression for the second oscillator as:

\[
M_2 = M_0 e^{-pt} \sin(\omega_c t) .
\] (28)

If we assume that the initial amplitudes of the first and third oscillators are equal, we can then write the expression for the signal as given in Eq. (19) as:

\[
M_x = M_0 e^{-pt} [1 + M e^{-rt} F(\theta)] \sin(\omega_c t)
\] (29)

where the ratio of the oscillator amplitudes, \( M \), is given by:

\[
M = \frac{2M_0}{M_0} .
\] (30)

At the magic angle, i.e., where \( \alpha = 0 \), Eq. (29) is seen to reduce to:

\[
M_x = M_0 e^{-pt} \sin(\omega_c t) .
\] (31)

Eq. (31) is seen to represent a single exponentially decaying sinusoid, which is what is experimentally observed.1

B. Data reduction. As we previously stated, \(^{131}\)Xe is not commercially available without a \(^{129}\)Xe impurity. For this reason, we chose to enhance the unwanted \(^{129}\)Xe signal by adding enriched \(^{129}\)Xe to the cell so that we could ensure its complete
removal from the data before we analyzed the decay of the $^{131}$Xe ensemble.

Typical raw data is displayed in Figure 3. This data was taken off the magic angle, thus the non-exponential character of the $^{131}$Xe decay is clearly observed. Since the nuclear spin of $^{129}$Xe is $1/2$, its decay at all cell orientations is simple exponential. The form of the $^{129}$Xe decay is thus exactly known and we then utilized a Kalman filtering technique to remove this signal from the raw data. The Kalman technique was used since this method was designed to remove 'signals' buried in noise. In this situation, we simply treat the $^{131}$Xe signal and the noise as 'noise' and remove a single exponentially decaying sinusoid representing the effects of $^{129}$Xe. The result of this processing is displayed in Figure 4. A 'fast fourier transform' was then performed on the data with the $^{129}$Xe signal removed to verify that the $^{129}$Xe had been completely removed.

Due to the subtle differences between data fitting with a Kalman technique and a Least Squares method$^{22}$, we chose to employ a Least Squares routine to fit the $^{131}$Xe data to our model. We used an eight parameter fitting routine of the form:

$$S = x_3 e^{-(x_4 + 2x_5)t} \left[ 1 + Me^{-x_6 t} F(\theta) \right] \sin (x_1 + x_2 t) + x_7 + x_8 t \quad (32)$$

$$F(\theta) = \begin{cases} \cos(x_6 t) + (x_5 / x_6) \sin (x_6 t) \\ \cosh (x_6 t) + (x_5 / x_6) \sinh (x_6 t) \end{cases} \quad (33a)$$

$$F(\theta) = \begin{cases} \cosh (x_6 t) + (x_5 / x_6) \sinh (x_6 t) \end{cases} \quad (33b)$$

Eq. (32) was used with Eq.'s (33a) and (33b) for the angle data far from the magic angle and very near the magic angle,
respectively. In Eq.'s (32 through 33b), the eight parameters represent the following terms:

\[ x_1 = \text{initial phases} \]  
\[ x_2 = \omega_c \]  
\[ x_3 = M^0_{xc} \]  
\[ x_4 = 1/T_2 \]  
\[ x_5 = 1/T_Q \]  
\[ x_6 = \left( \frac{1}{T_Q} \right)^2 - \left[ \frac{e^2 Q q}{4} \left( 3 \cos^2 \theta - 1 \right) \right]^2 \]  
\[ x_7 = \text{dc offset of the signal} \]  
\[ x_8 = \text{linear drift in the dc offset.} \]

The factor \( \tau_s/(\tau_s + \tau_v) \) in Eq. (34f) results from the time average of the dephasing terms over the duration of the collision. \( \tau_s \) is the duration of the interaction, that is the sticking time on the wall, and \( \tau_v \) is the time between collision.

In the fitting routine, we have set the initial phases of the three oscillators to be equal. There is no a priori reason why this should not be and we observe nothing in the results of our data fit to indicate that this is not a valid assumption.
We further had to make an assumption concerning the ratio of the oscillator amplitudes, M given by Eq. (30). An attempt to let this vary in the Least Squares routine resulted in instabilities in the data fit. In Ref. (1), we considered the case of low polarization. In this limit, using Eq. (8 and 10) of Ref. (1) and Eqs. (18a-c) we find:

$$M^O_{x1} : M^O_{x2} : M^O_{x3} = 3 : 4 : 3 \quad . \quad (35)$$

In the limit of complete polarization, on the other hand, which implies $\rho_{11}=1$ in Eq. (8) of Ref. (1), we find:

$$M^O_{x1} : M^O_{x2} : M^O_{x3} = 1 : 2 : 1 \quad . \quad (36)$$

We have good reason to believe that for these experiments, the nuclear ensemble was not in the low polarization limit, however, we also definitely know that the nuclear ensemble did not attain 100% polarization, this implied:

$$1 < M < 1.5 \quad \quad (37)$$

where 1 is the limit of complete polarization and 1.5 is the low polarization limit. We additionally demand that this ratio remain constant over all the angles at a given temperature and pumping time. This is, as was argued in Ref. (1), due to the fact that the first order quadrupolar interaction does not enter into the longitudinal relaxation of the noble gas ensemble and hence cannot affect the initial amplitude of the signal. By employing different ratios under the above constraints in the eight parameter fitting routine, we found that $M = 1.35$ gave the best results. This value was then fixed in the routine.
V. RESULTS

The three parameters of interest in the eight parameter
least squares routine are $x_4$, $x_5$ and $x_6$. From these terms we
derive the phenomenological relaxation rate,

$$\frac{1}{T_2} = (3.76 \pm .41) \times 10^{-3} \text{ sec}^{-1}$$

(38)

and the transverse quadrupole relaxation rate,

$$\frac{1}{T_Q} = (5.94 \pm .65) \times 10^{-4} \text{ sec}^{-1}$$

(39)

The angularly dependent dephasing rate, $\Delta a$, is displayed in
Figure 5. The errors associated with the derived terms represent
one standard deviation. The solid line in Figure 5 represents
a least squares fit of the time average of the dephasing rate to
the equation:

$$A(\theta) = \left( \frac{\gamma_s}{\tau_s + \tau_v} \right) \frac{e^2 Q q}{4} \left[ 3 \cos^2 \theta - 1 \right]$$

(40)

where $\theta$ is the angle between the magnetic field axis (z-axis)
and the imaginary axis diametrically through the tip of the cell.
From the fit we find:

$$A(\theta) = (8.9 \pm 0.4) \times 10^{-3} \left[ 3 \cos^2 \theta - 1 \right]$$

(41)

Comparing Eq. (40) and Eq. (41) we find:

$$\left( \frac{\gamma_s}{\tau_s + \tau_v} \right) e^2 Q q = 3.56 \times 10^{-2} \text{ sec}^{-1}$$

(42)
We can estimate the sticking time on the wall, \( \tau_s \), by assuming a simple adsorption model. The sticking time is then given by \(^24\):

\[
\tau_s = \frac{E_a}{kT} \exp \left( \frac{-E}{kT} \right)
\]

(43)

where \( \tau_o \) is taken to be \( 10^{-12} \) sec, and \( E_a \) is the adsorption energy. Taking \( E_a \) to be 0.13 eV\(^{13}\), we compute: \( \tau_s = 6.6 \times 10^{-11} \) sec. \( \tau_v \) is the time between wall collisions, and it can be estimated from simple gas kinetic arguments:\(^25\)

\[
\tau_v = \frac{4 \text{ Vol}}{v \text{ A}_s}
\]

(44)

where Vol is the volume of the cell, \( A_s \) is the surface area and \( v \) is the kinetic velocity of the xenon atoms. Evaluating Eq. (44) for our experimental conditions yields: \( \tau_v = 3.5 \times 10^{-5} \) sec. Since \( \tau_s \ll \tau_v \), we can neglect \( \tau_s \) in the denominator of Eq. (42) and calculate the average electric field gradient:

\[
eq = -3.45 \times 10^{11} \text{ statcoulombs cm}^{-3}
\]

(45)

Recently Keenan et al\(^{26}\) estimated the electric field gradient which arises at the nucleus of a \(^{131}\)Xe atom due to a distortion of the spherical shells. This distortion takes the form of electronic excitation with dipole selection rules. They assumed a one electron model and the excitation \( 5p^6 \rightarrow 5p^56s \). The field gradient was then estimated to be:

\[
eq = -7.0 \times 10^{12} \text{ statcoulombs cm}^{-3}
\]

(46)

Given the approximations, we find our value in good agreement with this estimate.
The transverse quadrupole rate is given in Ref. (5) as:

\[
\frac{1}{T_Q} = \frac{36\pi}{5} \frac{\tau s}{\tau s + \tau v} \left[ \frac{e^2 Q g}{3} \right]^2 \tau_c .
\]  

(47)

From our measured value of the quadrupole relaxation rate given in Eq. (39), and our estimate of the electric field gradient strength given in Eq. (45), we compute the correlation time for this interaction to be: \( \tau_c = 3.5 \times 10^{-7} \) sec.

Finally as a check on our parameter fit, we compare the phenomenological relaxation rate of the \(^{131}\text{Xe}\) with the decay rate of the \(^{129}\text{Xe}\) signal. From previous measurements\(^{13,14}\) on the decay of \(^{129}\text{Xe}\) and \(^{131}\text{Xe}\), we predict that at this temperature the predominant relaxation is due to spin exchange collision of the xenon atoms with the rubidium vapor. We then write:

\[
\frac{1}{T_2} = N_{\text{Rb}} \sigma_{\text{ex}} V_{\text{rel}}
\]  

(48)

where \(N_{\text{Rb}}\) is the rubidium density, \(\sigma_{\text{ex}}\) is the Rb-Xe spin exchange cross section and \(V_{\text{rel}}\) is the relative Rb-Xe velocity. The ratio of relaxation rates is simply the ratio of cross section for spin exchange. From the \(^{129}\text{Xe}\) signal, which we extracted from the original data, we found:

\[
1/T = \left(1.34 \pm 0.7\right) \times 10^{-2} \text{ sec}^{-1}
\]  

(49)

and thus we estimate the cross section ratio to be:

\[
\sigma_{^{129}\text{Xe}} / \sigma_{^{131}\text{Xe}} = 1/3.56 .
\]  

(50)
From Herman's model of Rb-noble-gas spin exchange we predict the ratio of cross sections to be:

$$\frac{\sigma_{129}^{ex}}{\sigma_{131}^{ex}} = \frac{1}{2.28} \quad (51)$$

We believe that this is very good agreement supporting the validity of our fitting procedure, and that the difference between Herman's prediction and our estimate is due to the contribution of the Rb-Xe van der Waals molecules in the alkali-noble-gas spin exchange interaction.\(^{14,28}\)

VI. DISCUSSION

We have investigated the nuclear transverse spin relaxation of \(^{131}\)Xe under a quadrupole coupling between the \(^{131}\)Xe nucleus and the electric field gradients which arise at the nucleus during collisions of the xenon atoms with the walls of the cell. We picture the collisions as resulting in a deformation of the xenon electronic shells. It is this deformation which produces the electric fields at the xenon nucleus. The angular dependence of this interaction is interpreted to mean that adsorption sites on the cell walls have a non-isotropic distribution. Further supporting this view is our finding of a dependence of this interaction on the distribution of the metallic rubidium on the cell walls. A nearly uniform distribution of the rubidium reservoir over the surface of the cell results in a very strong relaxation of the \(^{131}\)Xe ensemble, in fact complete relaxation is found to take place in a few precessional periods, and thus usable data cannot be obtained. In the cells that are uniformly coated with RbH, the adsorption sites on the walls appear to be isotropic with the location of excess rubidium metal being different from the rest. Relaxation data that fit to our model
are obtained only in those RbH-coated cells in which the rubidium metal is driven to the tip of the cell.

We believe that our technique of observing this effect is quite valuable since we obtain information from both the dephasing and transverse relaxation, which have different dependencies on the interaction parameters. We have, however, still not been able to definitely determine the adsorption energy or sticking time of the xenon atoms on the cell wall or the correlation time of the interaction, and thus a precise value of the electric field gradient, which we estimated, remains in some question. We have begun experiments to investigate these two parameters which will allow us to make a better determination of the interaction strength.

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8. L. I. Schiff, in Ref. 7, p. 247.


12. C. P. Slichter, in Ref. 11, p. 158.


2-28

15. Purchased from Monsanto Research Corp., Miamisburg, Ohio 45342.

16. One of us (TMK) has disclosed a patent on this technique.


22. A Kalman filter yields the best estimate of the parameter set at a given point based on all past measurements, whereas a Least Squares technique gives the best global estimate of the parameter set based on all measurements. In the limit that the model is exact the Kalman filter and the Least Squares method are identical. In our situation, the form of the $^{129}$Xe signal is known exactly, thus it is expedient to remove its effects with the Kalman filter. Due to the estimates used in arriving at the form of the $^{131}$Xe signal, we chose to use the Least Squares technique and estimate the parameters on the global sense of the data.


FIGURE CAPTIONS

Figure 1. Schematic representation of the experimental apparatus

Figure 2. Geometrical arrangements of the experimental cell

Figure 3. Typical example of the raw signal. Low frequency component is due to the $^{131}$Xe precession, while the high frequency is the $^{129}$Xe signal. This data was taken at the cell orientation at which the magnetic field axis coincides with the imaginary axis diametrically through the tip of the spherical cell.

Figure 4. Typical example of the signal after the effects of the $^{129}$Xe have been removed. The data shown in Figure 2-3 are displayed.

Figure 5. Quadrupole dephasing terms as a function of angle. The error bars on the data points represent one standard deviation and the solid line is a Least Squares fit to the data.
Figure 1
Figure 3
Figure 4

RELATIVE SIGNAL

TIME (sec)
Figure 5
SECTION III

NOBLE GAS NUCLEAR FREQUENCY SHIFT

3.1 INTRODUCTION

The shift of the precessional frequency of the noble gas nucleus in a magnetic field reported in this section is associated with the transfer of angular momentum from an electronically polarized alkali atom to an unpolarized noble gas nucleus in the spin exchange process. Such frequency shift has been reported for the case of free electrons interacting with optically pumped alkali atoms.¹⁻³. No such information is yet available to our knowledge for noble gas nuclei, such as Kr⁸³, Xe¹²⁹ and Xe¹³¹.

Since the angular rate in the nuclear magnetic resonance (NMR) gyro is sensed as a shift in the precession frequency of certain nuclear species, the information regarding the frequency shift can lead to a determination of gyro bias shift due to the spin exchange interaction with the polarized alkali-vapor. As will be shown later, the frequency shift of the noble gas nucleus is strongly dependent on both the number density of the alkali-atom and the alkali polarization. This would mean that the same information would also lead to determinations of temperature dependence and the alkali-pumping light intensity dependence of the NMR gyro bias.

In this section we review briefly the theoretical background of the spin exchange interaction to give an understanding of the frequency shift, and subsequently present the experimental results.
3.2 THEORY OF FREQUENCY SHIFT

In order to review the theory, we will rely on the results previously reported\(^4\), and repeat only selected details here as clarity demands.

We start with Herman's effective interaction Hamiltonian between an alkali atom and noble gas nucleus in the form:\(^5\)

\[
\mathcal{H}_{\text{eff}} = \left( \frac{16\pi}{3} \right) g_N \mu_N \mu_0 \mu_1(B) \right) \eta^2 \vec{I} \cdot \vec{S} \tag{52}
\]

where \(\vec{I}\) is the noble-gas nuclear spin, \(\vec{S}\) the alkali electronic spin, \(g_N\) the nuclear g-factor, \(\mu_N\) the nuclear magneton, and \(\mu_0\) the Bohr magneton. \(\eta\) is the exchange enhancement factor, given by:

\[
\eta(R) = \left[ 1 - \sum_{i=2}^{n} \frac{<i|1> u_i(B)}{u_1(B)} \right] \tag{53}
\]

where \(R\) is the internuclear separation between the alkali and noble gas atoms. The summation over \(i\) refers to the alkali valence electron (1) and the noble gas electrons (2,3...,\(n\)). The factors \(<i|1>\) are the overlap integrals and \(u_1(B)\) is the amplitude of the ith orbital of the isolated atom at the noble gas nucleus, B. For the internuclear separation of \(R = 7.8a_0\) (\(a_0\) is the unit of atomic distance; \(a_0 = 0.529X10^{-8}\)cm), the values of \(\eta\) for the Rb-Kr and Rb-Xe system have been calculated to be\(^4\):

\[
\eta_{\text{Kr}} = 40 \text{ and } \eta_{\text{Xe}} = -59 \tag{54}
\]
We consider a gaseous mixture of alkali and noble gas atoms subjected from the outside only to a constant uniform magnetic field, but interacting with each other through the relatively short range forces, so that the otherwise free atoms occasionally collide with each other. These collisions may be either of the binary type or molecular forming (three body) type. All collisions are such that the mean time between collisions, $\tau_A$, is long compared to the duration of the collision, $\tau_D$. It is further assumed that the external magnetic field is so weak that the precession frequencies of the electron moment and the nuclear moment are orders of magnitude smaller than the strength of the interaction given by Eq. (52), when the internuclear separation is within the atomic range.

In what follows we take the total Hamiltonian acting on the spin variables to be:

$$\mathcal{H} = -h\omega_S S_z - h\omega_I I_z + [h\gamma(R) \vec{I} \cdot \vec{S}]$$  \hspace{1cm} (55)

where the first two terms of the right hand side of the equation are the coupling to the external magnetic field, which will be represented by $\mathcal{H}_0$, and the last term represents the exchange interaction, where $\gamma(R)$ is the strength of the interaction:

$$\gamma(R) = \left(\frac{16\pi}{3}\right) g_N \mu_N g_{\text{u}}(B)^2 \eta(R)^2/h.$$  \hspace{1cm} (56)

$\gamma(R)$ falls off rapidly with internuclear separation, $R$, and thus the kinematics of the interaction is to picture the spins before a collision as acted on by $\mathcal{H}_0$, then coming under the combined action of $\mathcal{H}_0 + h\gamma(R) \vec{I} \cdot \vec{S}$ during the interaction and finally, as the particles separate, going back to the uniform precession of $\mathcal{H}_0$. 

3-3
We now consider a much simplified system consisting of an atom with a single electron in a $2S_{1/2}$ state and no nuclear spin (the alkali atom $S=1/2$) and an atom with no electronic spin, $S'=0$ and nuclear spin $I=1/2$ (the noble gas atom). The system is described quantum mechanically by a density matrix of $(2S + 1)(2I + 1)$ dimensions, namely, by a $4 \times 4$ matrix.

The density matrix can be constructed in the representation of the ortho-normal set of eigenfunctions of $I^2$, $I_z$, and $S^2$, $S_z$ for the free (noninteracting) atoms. The time development of the density matrix resulting solely from the spin exchange interaction can then be investigated in the interaction representation.

The origin of the shift of the noble gas nuclear precession frequency involves the equation of motion for the mean value of the operator $I^+$ (or $I^-$), where, as commonly defined, $I^± = I_x ± i I_y$, in the Schrödinger representation. Following the work of Major\textsuperscript{6}, it has been shown that

$$\frac{d}{dt} <I^+> = - \frac{1}{\tau_A} \left[ (\frac{1}{2}) \gamma^2 \frac{\tau^2}{\text{ave} D} - i <S_2> \frac{\gamma \text{ave} \gamma D}{1 + \gamma^2 \frac{\tau^2}{\text{ave} D}} \right] <I^+> (57)$$

$$- \frac{1}{\tau_2} <I^+>$$

where $\gamma_{\text{ave}}$ is the strength of the spin exchange interaction, given by Eq. (56), averaged over the collisional parameters.
The real part of the coefficient of $<I^+>$ represents the phase coherence decay time, $T_{ex}$, due to the spin-exchange interaction, i.e.,

$$T_{ex}^{-1} = \frac{1}{2} \frac{1}{\tau_A} \frac{\gamma_{ave}^2 \tau_D^2}{1 + \gamma_{ave}^2 \tau_D^2}.$$  \hspace{1cm} (58)

The imaginary term, which is seen to be proportional to the electron spin polarization, represents a shift in the precession frequency of the nuclear spin in the external magnetic field:

$$\Delta \omega = \left( \frac{1}{\tau_A} \right) \frac{\gamma_{ave} \tau_D}{1 + \gamma_{ave}^2 \tau_D^2} <S_z>.$$ \hspace{1cm} (59)

This shift can be written as an equivalent magnetic field, given by:

$$\Delta H = \left( \frac{1}{g_N \mu_N} \right) \left( \frac{1}{\tau_A} \right) \frac{\gamma_{ave} \tau_D}{1 + \gamma_{ave}^2 \tau_D^2} <S_z>.$$ \hspace{1cm} (60)

where $\Delta H$ is now a magnetic field 'experienced' by the noble gas atoms due to the presence of the alkali atom.

3.2.1 Frequency Shift Due to Exchange Process of the Binary Type

Consider the spin exchange process due to the binary type collisions, for which $\gamma_{ave} \tau_D \ll 1^7$. Eq. (60) can now be written as:

$$\Delta H = \left( \frac{1}{g_N \mu_N} \right) \left( \frac{1}{\tau_A} \right) \gamma_{ave} \tau_D <S_z>.$$ \hspace{1cm} (61)
The collision rate can be written as:

\[ \tau_D = \frac{\bar{d}^2 N_A v_{rel}}{} \]  \hspace{1cm} (62)

where \( \bar{d} \) is the mean interaction distance and \( N_A \) is the alkali atom number density. \( v_{rel} \) is the mean relative velocity between the alkali atoms and noble-gas atoms. The binary collision duration is taken to be:

\[ \tau_D = \frac{\bar{d}}{v_{rel}} \] \hspace{1cm} (63)

Combining Eq's. (62) and (63) with the definition of \( \gamma \) given in Eq. (56), Eq. (61) is found to be:

\[ \Delta H = \frac{16\pi^2}{3} \bar{d}^3 N_A \mu_o \left( \frac{u_1(B)}{2} \right)^2 \left( \eta(R) \right)^2 \langle S_z \rangle \] \hspace{1cm} (64)

where we have appropriately averaged values of \( u_1(B) \) and \( \eta(R) \). Since \( u_1(B) \) is the wave function of the unperturbed alkali valence electronic orbital, \( 2\pi\bar{d}^3 \left( \frac{u_1(B)}{2} \right)^2 \) is of order unity.

We note in Eq. (64) that the value of \( \Delta H \) for different isotopes of the same noble gas atom is the same for the same alkali polarization environment (neglecting a small effect of the different nuclear structure), but that different noble gas species experience different magnetic field shifts, mainly due to the different values of the exchange enhancement factor, \( \eta \).
If we pick $<S_z> \sim 0.1$ and a sample temperature of $65^\circ$C, we find the magnetic field shifts for Kr and Xe due to the presence of Rb as:

$$\Delta H_{Kr} \sim 2.6 \mu G, \text{ and}$$

$$\Delta H_{Xe} \sim 5.7 \mu G .$$

Under the present mechanization of Litton's NMR gyro, the gyro bias shift, $\Omega_a'$ due to the magnetic fields seen by the noble gas atoms is shown to be:

$$\Omega_a = \frac{\gamma_b}{\gamma_a - 1} (\Delta H_a - \Delta H_b)$$

(66)

where the subscripts $a$ and $b$ refer to the two nuclear species to be used in the sample, and the $\gamma$'s are the corresponding nuclear gyro magnetic ratios. When the results of Eq. (65) are used, the gyro bias shift becomes 580 deg/hr. Samples that contain a pair of isotopic nuclei of the same noble gas atom, such as Xe$^{129}$ and Xe$^{131}$, will show essentially zero bias shift since the factor $(\Delta H_a - \Delta H_b)$ is diminished.

In an NMR gyro, this gyro bias shift should not be a problem as long as it remained constant. Eq. (64) indicates, however, the gyro bias shift will vary as a function of the cell temperature (temperature sensitivity) or of the alkali-pumping light intensity. These shifts can be clearly deduced by noting that a change of the cell temperature will result in a change of both
the density of alkali atoms, \( N_A \), and the alkali-polarization, \( \langle S_z \rangle \), and that a variation of the pumping light intensity at a fixed temperature will result in a variation of \( \langle S_z \rangle \). It is for this reason that two nuclear isotopes of the same noble gas atoms, i.e. Xe\(^{129}\) and Xe\(^{131}\), are the preferred embodiment in the Litton approach to the development of an NMR gyro.

3.2.2 Frequency Shift in a Pair of Isotopic Nuclei of the Same Noble Gas

We have argued in the previous section that, when the spin exchange is dominated by binary type collisions, a pair of isotopic nuclei of the same noble gas atom experiences the same frequency shifts, which results in a "zero" gyro bias shift. Interactions other than the dipolar spin-exchange process that may lead to a frequency shift are discussed in the following.

Nuclear Structure Effect

There will be a limit to the condition that a pair of isotopic nuclei of the same noble gas atom experiences the same frequency shift. This is due to the isotopic "hyperfine anomaly" resulting from nuclear structure effects, which makes the strength of the hyperfine interaction, \( \gamma(R) \), not strictly the same for each member of the pair. Nevertheless, the departure due to this effect of the small variance in the gyromagnetic ratios of the pair\(^6\) is never more than one part in \( 10^3 \).

Effect of Molecular Formation

It has been shown previously\(^7,9\) that, in addition to the binary collisions, the spin exchange takes place through the formation of weakly-bound molecules, such as Rb-Xe, in the presence of a
third colliding body, such as $N_2$ buffer gas (three body collisions). If the duration of the interaction is taken to be the duration of the three body collision, $\tau_c$, the magnetic field experienced by the noble gas nuclei due to the formation of molecules, $\Delta H_{\text{mole}}$, is shown to be:

$$\Delta H_{\text{mole}} = \frac{1}{q_N \mu N} \left( \frac{1}{T_F} \right) \frac{\gamma \text{ave} \tau_c}{1 + \gamma^2 \text{ave} \tau_c^2} <S_z>$$

(67)

where $T_F$ is the molecular formation rate, and both $T_F$ and $\tau_c$ are functions of the buffer gas density. It remains to be investigated if the interaction strength and the collision time are the same for different isotopes of the same noble gas under conditions of identical buffer gas density.

**Effect of Nuclear Quadrupole Moment**

Consider the nucleus of Xe$^{131}$ ($\bar{I} = 3/2$), the nuclear quadrupole moment of which interacts with the electric field gradient at the nuclear site. It has been stated in Section II that the electric field gradient arises from deformation of the otherwise spherical electronic shells of the xenon atom during inelastic collisions with the cell walls. (The quadrupole interaction is absent in the nucleus of Xe$^{129}$, because $\bar{I} = 1/2$.) The quadrupole interaction of the noble gas nucleus is modeled as $2I$ oscillators, namely $3$ oscillators for Xe$^{131}$, each with a single frequency. The frequencies are dependent on the orientation of the cell with respect to the magnetic field. When a cell, where the nuclear
quadrupole interaction is made cylindrically symmetric, is oriented to the 'magic' angle, the three oscillator frequencies become nearly identical to the frequency, $\omega_c$, given by Eq. (23a):

$$\omega_c = \omega + \frac{54b^2 \alpha^2}{\omega} \tag{68a}$$

where

$$\omega = -\gamma \hbar H \tag{68b}$$

$$b = \sin\theta \cos\theta \tag{68c}$$

where $\omega$ is the unperturbed nuclear precession frequency in a magnetic field $H$, and $\theta$ is the angle between the magnetic field axis and the axis of cylindrical symmetry. The strength of the quadrupole interaction, $\alpha$, has been shown to be:

$$\alpha = \left(\frac{\tau_s}{\tau_s + \tau_v}\right) \frac{e^2 \alpha}{4I(2I-1)} \tag{69}$$

where $e$ and $\alpha$ are the electric field gradient and the nuclear quadrupole moment, respectively, as described in Section II. $\tau_s$ is the sticking time of xenon atoms on the cell walls, and $\tau_v$ is the time between wall collisions.

Any departure of the nuclear precessional frequency, $\omega_c$, from the unperturbed frequency, $\omega$, results in the frequency shift. The shift is seen from Eq. (68a) to be proportional to the square of the strength of the nuclear quadrupole interaction. In a high magnetic field ($\omega$ is large), however, this frequency shift could be made relatively small. Discussions so far have been limited to the first order effect of the nuclear quadrupole interaction. The second order effects are small and are neglected.
3.3 EXPERIMENT

3.3.1 Description of Experiment

Shown in figure 6 is the experimental arrangement designed to measure the magnetic field experienced by the noble gas nucleus due to the spin exchange interaction with an optically oriented alkali vapor. This setup has been described in detail elsewhere\(^7\). Modifications of the arrangement are made so that the alkali pumping light beam can be made left or right hand circularly polarized by placing suitable polarizers in front of the light source. The left and right hand circular polarizers are made by laminating a sheet of plastic linear polarizer and two sheets of plastic retarder. The two polarizers so made transmit nearly equal intensity of rubidium D\(_1\) light. The amount of circularly polarized light was measured to be 92% for one and 96% for the other of the transmitted light.

The experiments were performed by, first, polarizing the noble gas nuclear spins through spin exchange collisions with the optically oriented rubidium vapor (Pump Mode). The rubidium atoms were oriented by the absorption of left or right hand circularly polarized rubidium D\(_1\) light in a standard longitudinal pumping scheme: This configuration is shown in figure 7(a). The pumping takes place in the presence of a longitudinal magnetic field (Pump Field) of approximately 100 mG along the z-axis. The longitudinal noble gas polarization, \(I_z\), has been shown to obey the usual pumping law, given the initial condition

\[
I_z(t=0) = I_z^0
\]

\[
\langle I_z(t) \rangle = \langle I_z^0 \rangle \{1-\exp(-t/T_p)\} \quad (70)
\]
Figure 6. Schematic representation of the experimental apparatus

NOTE: THIS FIGURE IS DIFFERENT FROM FIGURE 1 IN THEIR AXIS DEFINITIONS.
Figure 7. Magnetic field arrangement for (a) the longitudinal pumping of the noble gas nuclear spin polarization, (b) modulation of π/2 pulse, and (c) the precession of the polarized nuclear spins. For brevity, only one species of noble gas nuclear moment is shown (thick arrow).
where $T_p$ is the "pumping" time of the ensemble of noble gas nuclear spins. The ensemble was pumped for a time sufficient to build up a significant polarization. This time can range from 5 minutes at low sample temperatures to 30 minutes at high sample temperatures.

When a sufficient nuclear polarization was built up along the light beam axis (z-axis), the pump field was turned off, and, at the same time, a "classic" $\pi/2$ pulse was applied along the x-axis to the nuclear system. This "steers" the nuclear polarization away from the z-axis into the x-y plane as shown in figure 7(b).

Finally, the precession of the noble gas nuclear spin polarization about the z-axis was accomplished by applying a precession field ($\sim 100 \mu G$) along the z-axis, figure 7(c). The optically oriented rubidium vapor in this scheme, then, acts either to directly increase or decrease the effective precession field as experienced by the noble gas ensemble, depending on the circular polarizers used in the measurement.

The precession frequency of the ensemble of the noble gas nuclei is, correspondingly, increased or decreased such that:

$$\omega_1 = -\gamma (H_0 + \Delta H)$$

$$\omega_2 = -\gamma (H_0 - \Delta H)$$

(71)

where $H_0$ is the magnetic field applied externally along the precession axis and $\Delta H$ is the field experienced by the noble gas.
ensemble due to the spin exchange interaction. $\gamma$ is the nuclear gyromagnetic ratio of the noble gas atoms. The subscripts, 1 and 2, refer to the two different circular polarizers. From these, $\Delta H$ is readily obtained by measuring the precession frequencies, $\omega_1$ and $\omega_2$. A similar experimental arrangement has been employed previously by Balling, et al., for the measurement of frequency shifts of electrons interacting with the polarized alkali vapor through the spin exchange process.

In order to observe the precessing nuclear spin polarization signal, the rubidium vapor was mechanized as a magnetometer. The magnetometer detects magnetic field variations along the x-axis due to the precessing noble gas nuclear ensemble. The form of the observed signal has been shown to be $^7$:

$$S_i = S_a \exp \left[ -t/(T_i a) \right] \sin(\omega_{ai} + \phi_{ai})$$
$$+ S_b \exp \left[ -t/(T_i b) \right] \sin(\omega_{bi} + \phi_{bi}), \text{ for } i=1 \text{ and } 2$$

(72)

where the subscripts a and b refer to each species of noble gas nuclear isotope present in the cell, and i refers to the condition that either a left or a right hand circularly polarized beam is used in the experiment. $T_i$'s are the transverse relaxation times and $\phi$'s are the initial phases. Data are then analyzed utilizing a Kalman filter technique to obtain the nuclear precession frequencies.

The samples used in the measurements are all 1-ml spherical cells made of pyrex glass. The glass envelopes were baked overnight at 300°C under high vacuum. After the bake, rubidium metal was
distilled into the cell, and, a pair of noble gas isotopes, Xe$^{129}$ and Kr$^{83}$, or Xe$^{129}$ and Xe$^{131}$, together with buffer gases such as N$_2$ and/or He, were filled into the cell. In order to minimize the uncertainties of the gas pressures in the sealed-off cells, each of the spherical cells was completely covered with thermal insulation while the cell stems were sealed with a working flame. The rubidium metal in the sealed cells was then carefully driven into the small region of the cell stem area. The cells so made were then "aged" in an oven at 80°C for no less than two weeks before the measurements.

3.3.2 Experimental Results and Discussions

The results are presented for the three different sample cells listed in Table I. Cells 1-44 and 1-48 are for the Xe$^{129}$-Kr$^{83}$ pair and cell 1-75 is for the Xe$^{129}$-Xe$^{131}$ pair.

In figure 8, the magnetic fields, ΔH, experienced by Xe$^{129}$ and Kr$^{83}$ in cells 1-44 and 1-48 due to the spin exchange interaction with the optically oriented rubidium vapor are shown as a function of the cell temperature. The functional form of temperature dependence of the magnetic field qualitatively confirms the theory discussed earlier. As the cell temperature increases, the rubidium number density, $N_A$, increases, and, at the same time, the rubidium polarization, $<S_z>$, decreases, with ΔH being a function of the product of the two, $N_A<S_z>$. The magnitude of ΔH experienced by Xe$^{129}$ is larger than that by Kr$^{83}$ at all temperatures in the same cell (i.e. the same rubidium polarization environment). This is the result of the different magnitudes of the enhancement factors (i.e. $|\eta_{Xe}| > |\eta_{Kr}|$).
<table>
<thead>
<tr>
<th>Cell S/N</th>
<th>Alkali Metal</th>
<th>Nuclear Species</th>
<th>Buffer</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 - 44</td>
<td>Rb$^{87}$</td>
<td>0.2 Torr of Xe$^{129}$ and 5.0 Torr of Kr$^{83}$</td>
<td>15 Torr of N$_2$ and 140 Torr of He</td>
</tr>
<tr>
<td>1 - 48</td>
<td>Natural Rb</td>
<td>0.2 Torr of Xe$^{129}$ and 5.0 Torr of Kr$^{83}$</td>
<td>100 Torr of N$_2$</td>
</tr>
<tr>
<td>1 - 75</td>
<td>Natural Rb</td>
<td>0.1 Torr of Xe$^{129}$ and 0.4 Torr of Xe$^{131}$</td>
<td>10 Torr of N$_2$ and 100 Torr of He</td>
</tr>
</tbody>
</table>
Figure 8. The magnetic fields experienced by $\text{Xe}^{129}$ (solid square) and $\text{Kr}^{83}$ (empty square) nuclei in two different sample cells as a function of cell temperature.
Comparing the magnitude of $\Delta H$ experienced by each isotope in the two different cells, the ones in cell 1-48 are larger than those in cell 1-44. This is understood to mean that higher $N_2$ pressure (100 torr) in cell 1-48 provides a higher degree of rubidium polarization than that provided in cell 1-44 (15 torr of $N_2$).

Attention is directed in figure 8 to the fact that the ratio of the magnetic field experienced by $Xe^{129}$ to that by $Kr^{83}$ due to the spin exchange is insensitive to the cell temperatures. The same ratio is observed within the experimental uncertainty even in the two different cells. This is an expected result from Eq. (64). The ratio determined by this measurement is $2.2 \pm 0.1$.

Essentially the same features are observed for cell 1-75, which is for the $Xe^{129}$ and $Xe^{131}$ pair, as shown in figure 9. The magnetic fields, $\Delta H$, experienced by the two xenon isotopes are nearly identical at all temperatures. This confirms experimentally the theoretical argument presented earlier that, under the same alkali polarization environment, a pair of nuclear isotopes of the same noble gas would see nearly equal magnitudes of magnetic fields due to spin exchange with the optically oriented alkali vapor. The degree of the alkali polarization in cell 1-75 may be close to that in cell 1-44, in view of the fact that the magnitude of the magnetic fields experienced by $Xe^{129}$ is nearly the same in the two cells.

The same data shown in figures 8 and 9 are presented in figures 10 and 11, respectively, in terms of NMR gyro bias shift as a function of temperature. Figure 10 shows $Kr^{83}$-$Xe^{129}$ cell bias shifts of -60 deg/hr and -100 deg/hr for a $1^\circ C$ variation of the cell temperature at a nominal cell operating temperature of $75^\circ C$ (temperature sensitivity).
Figure 9. The magnetic fields experienced by both Xe$^{129}$ and Xe$^{131}$ nuclei as a function of the sample cell temperature. The difference of the fields experienced by the two nuclear species are too small to be clearly seen in the plot.
Figure 10. NMR gyro bias versus the cell temperature for the cells with the Xe$^{129}$ and Kr$^{83}$ pair of isotopes. The same data shown in figure 3-3 are used.
Figure 11. NMR gyro bias versus the cell temperature for the cell (1-75) with the Xe\textsuperscript{129} and Xe\textsuperscript{131} pair of isotopes. The same data shown in figure 3-4 are used. The large error bars are due to the computational error.
As shown in figure 11, the gyro bias shift in the cell with the $^{129}$Xe and $^{131}$Xe pair (cell 1-75) is at least an order of magnitude smaller than that in the cells with the $^{129}$Xe and Kr$^{83}$ pair. The large error bars in figure 11 indicate the computation error. (Two nearly equal numbers have to be subtracted to get the gyro bias.) The existence of any gyro bias shift in this cell is not surprising in view of the discussions mentioned previously. (A few percent of this bias shift could have been caused by the different polarizabilities of the two circular polarizers used in the experiment.) Notice that, at 78°C, the bias becomes maximum, i.e., when the cell is operated at this temperature, sensitivity of the gyro bias will be identically zero.

3.4 SUMMARY AND FUTURE PLANS

A theory of nuclear precession frequency shift due to the spin exchange interaction with an optically oriented vapor was reviewed. Experimental results are qualitatively in good agreement with the theory. In particular, it has been confirmed experimentally that a pair of nuclear isotopes of the same noble gas atoms experience nearly equal frequency shifts under an identical alkali polarization environment.

The gyro bias shift in the cell with the $^{129}$Xe and $^{131}$Xe pair is at least an order of magnitude smaller than that of the cell with the $^{129}$Xe and Kr$^{83}$ pair. It has been found in this work that there is a certain cell temperature, at which the temperature sensitivity of the gyro bias shift is identically zero. In order to possibly utilize the point of zero temperature sensitivity in an NMR gyro, the existence of any gyro bias shift
would have to be investigated further within the frame of the responsible physical phenomena discussed in the theoretical section. Some of the possible causes could be studied advantageously by varying the temperature of the cell wall while maintaining the product of alkali number density and the alkali polarization constant. This could be done in an experimental arrangement in which the wall and the cell tip (where the alkali metal reservoir is confined) temperatures are controlled separately.
3.5 BIBLIOGRAPHY FOR SECTION III

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