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TRANSFER OF NUCLEAR ALIGNMENT COHERENCE IN ^{131}Xe DURING ELECTRON EXCHANGE COLLISIONS

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April 22, 1968

Transfer of Nuclear Alignment Coherence in ^{131}Xe During Electron Exchange Collisions*

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Experimental evidence of the transfer of the transverse component of the nuclear alignment in the electron-exchange collisions between the $^1S_0$ ground state and $^3P_2$ metastable state of ^{131}Xe (I = 3/2) is described. In this experiment, the nucleus in the atomic ground state, $^1S_0$, was aligned through metastability-exchange collisions with aligned metastable-state atoms which were formed by electron impact excitation. The coherence among the magnetic sublevels of the nucleus was then created by nuclear magnetic resonance (nmr) at 200 to 2000 Hz. A second collision between the $^1S_0$ ground state and the $^3P_2$ metastable state transfers the nuclear coherence to the metastable state and results in the modulation of the light absorbed by the metastable state at the characteristic nmr frequency.

Recently, Partridge and Series[1] reported that the transverse component of the nuclear magnetization of ^3He can be transferred to the $^3S_1$ metastable state of ^3He through metastability exchange collisions. A very

clever theoretical technique developed by Partridge and Series gives excellent agreement with the observed phenomena. In their experiment, since I = 1/2 for ^{3}He, it was necessary to orient the nucleus first. This was accomplished through metastability-exchange collisions with the oriented metastable state which had been pumped optically by the methods demonstrated by Walter, Colegrove, and Shearer.[2]

In this letter we report the experimental evidence that the coherence can be transferred also in the case of nuclear alignment. We used ^{131}Xe to test the idea, since this isotope has a nuclear spin I = 3/2, which may be aligned. The natural abundance of the ^{131}Xe isotope is 21%, so ordinary Xenon was employed. The test of the transfer of coherence in collisions, whether the nucleus is aligned or oriented, is made in terms of whether the resonance absorption by the metastable state shows the characteristic nmr frequency modulation. Also, such phenomena are not limited to the metastability-exchange collisions, but occur also in collisions in general.

It was demonstrated by Ruff and Carver[3] that in spin-exchange collisions between the Na and H, both being in the ground state, the coherence created in the hydrogen ground state can be transferred to the sodium ground state, resulting in the modulation, at the hydrogen resonance frequency in the transverse beam, of light absorbed by sodium atoms.

The experiments of Partridge and Series[1] and Ruff and Carver can be explained in terms of a classical model similar to that involved in the derivation of Block's equation; in fact, one should be able to observe

the modulated signal by the classical magnetic induction method. However, there is no simple classical analog associated with the experiment we performed, and it is not possible to observe the signal by use of the nmr induction method.

The mechanism involved in the experiment can be explained in the following way. First of all, our previous experiment[4] showed (from the observation of paramagnetic resonance) that the metastable state of ^{131}Xe is aligned by the electron impact excitation. This means the hyperfine structure states of F = 7/2, 5/2, and 3/2 are all aligned by electron impact. With this information and from the paramagnetic resonance of the even isotopes (I = 0 and no hfs), we can easily deduce that the nuclear spin is aligned through the hyperfine interaction.

In the decoupled representation, the partial-density matrix representing the metastable state F, $\rho_{m_F m_F'}(F)$, immediately after the collision, is given by

$$\rho_{m_F m_F'}(F) = [U\rho(e)\otimes\rho(n)U^{\dagger}]_{m_F m_F'} \quad ,$$

where $\rho(e)$ is the electronic part of the density matrix of the metastable state, and $\rho(n)$ is the nuclear part of the density matrix of the metastable state. Because of the decoupled representation, U is the unitary transformation operator given by $\{\psi_{F,m_F}\} = U(\psi_{I,m_I} \otimes \psi_{J,m_J})$. If we consider the metastability-exchange collisions between the ^1S_o ground state and $^3P_2$ metastable state, the aligned nucleus in the metastable state now becomes the aligned nucleus in the newly formed ^1S_o ground state. The nuclear spin direction of the newly formed metastable state is completely

random, so there results a net reduction of the alignment of the metastable state. Since the spin relaxation time of the nucleus in the ground state is very long compared with its spin relaxation time in the metastable state, it is easy to see that after many collisions of the type mentioned above the ground state atoms will be aligned to the same degree as the metastable state atoms. Now, if we cause nuclear magnetic resonance of the $^1S_0$ ground state, there will be coherence among the magnetic sublevels of the ground state. If we denote the density matrix operator of the ground state by $\rho(g)$, $\rho_{m_J m_I'}(g) \neq 0$ for $m_I = m_I'$ as well as for $m_I = m_I'$. Furthermore, the off-diagonal component of $\rho_{m_I m_I'}(g)$ oscillates at the characteristic nmr frequency and at its harmonics if $I > 1/2$. Therefore, if we now consider the newly formed metastable state, $\rho_{m_F m_F'}(F) = [U\rho(e) \otimes \rho(g)U^\dagger]_{m_F m_F'}$, in which $\rho(g)$ oscillates with time, $\rho_{m_F m_F'}(F)$ will also oscillate at the characteristic nmr frequency. Hence, the light absorbed by the metastable-state atoms shows the characteristic nuclear magnetic resonance frequency modulation which is three orders of magnitude lower than the paramagnetic resonance frequency of the metastable state.

The equations that describe the above-mentioned mechanism can be written as

$$\frac{d}{dt}\rho(g) = \frac{i}{\hbar}\left[\{\mathcal{H}_o + \gamma I_z H + \gamma H_1(I_x \cos\omega t + I_y \sin\omega t)\ ,\ \rho(g)\right] - \Gamma_g\,\rho(g) + R_g\,\rho(n)\ , \tag{1}$$

$$\frac{d\rho(F)}{dt} = -\frac{i}{\hbar}\left[(\mathcal{H}_o + \gamma F_z),\ \rho(F)\right] - \Gamma_\mu \rho(F) + \frac{\rho^\circ(F)}{T_p} + R_\mu[U\rho(e)\otimes\rho(g)U^\dagger], \tag{2}$$

where Γ_g, Γ_μ are the damping constant of the ground state and the metastable state respectively, R_g and R_μ are the production rate of the ground state and metastable state through the electron exchange collisions, and $\frac{\rho^\circ(F)}{T_p}$ is the production rate of the metastable state by electron impact excitation. Equation (1) is identical to that of Partridge and Series,[1] and Eq. (2) is simpler than their expression because we do not have the optical pumping term. Solving Eq. (1) and Eq. (2) simultaneously, one can obtain the expression for the light absorbed by the metastable state as $I_{ab} = K \operatorname{tr} \rho M$, where M is the electric dipole monitoring operator.

I_{ab} is given in the form

$$I_{ab} \propto \frac{\left[4\sqrt{3\alpha}\,\{\rho_{-1/2\ -3/2}(F)-\rho_{3/2\ 1/2}(F)\}+2\sqrt{3\gamma}\,\{\rho_{-1/2\ +3/2}(F)-\rho_{3/2\ 1/2}(F)\}\right]}{[\Gamma_\mu+i(g_F\omega_\ell-\omega_o)]} + \frac{\left[4\sqrt{3\alpha}\,\{\rho_{-3/2\ -1/2}(F)-\rho_{1/2\ 3/2}(F)\}+2\sqrt{3\gamma}\,\{\rho_{-3/2\ -1/2}(F)-\rho_{1/2\ 3/2}(F)\}\right]}{[\Gamma_\mu-i(g_F\omega_\ell-\omega_o)]} \qquad (3)$$

for the fundamental modulation frequency of the F = 3/2 state, when the applied static field is $H\hat{k}$, the oscillating field is H_1 ($\hat{i}$ cos ωt + $\hat{j}$ sin ωt), and the optical absorption by ^{131}Xe (in the $^3P_2$, F = 3/2 state) is detected with the <u>linear</u> polarizer set to select $\hat{x} + \hat{k}$ linearly polarized light; α and γ are the production rates of $\rho^{(e)}_{m_J} = 2,\ m_J = 2$ and $\rho^{(e)}_{m_J} = 0,\ m_J = 0$.[4] Results are similar for F = 7/2 and for F = 5/2. By means of perturbation theory,[5] $\rho^{(F)}_{m_F,m_F}$ can be evaluated in a straightforward manner in terms of a linear expansion of $1/[\Gamma_g + i(\Delta n)\ p]$, since

$$\rho^{(g)}_{m_I m_I'} = e^{-i(m_I - m_I')\omega_o t} \times \sum_{n,n',m_I,m_I''} \frac{R_g\, D^I_{m_I n}(\beta)\, D^I_{n m_I''}(\beta)\, D^I_{m_I' n'}(\beta)\, D_{n' m_I'}(\beta)\, \rho^o_{m_I'' m_I''}}{\Gamma_g + i(n-n')p} \tag{4}$$

from the theory of magnetic resonance,[6] where $\rho^o_{m_I'' m_I''}$ is the initial population of the m_I'' due to the transfer of the longitudinal component of the metastable state alignment; β is the angle that the effective field in the rotating coordinate system makes with $H\hat{k}$, and is given by $\beta = \tan^{-1} \frac{\gamma H_1}{\gamma(H-H_o)} = \tan^{-1} \frac{b}{\delta}$ in standard notation. Thus, if $\rho_{m_F m_F'}(F)$ is evaluated, the form of the modulated signal is given by

$$I_{ab} \propto \frac{1}{\Gamma_\mu} \left\{ \left(\frac{A}{\Gamma_g} + \frac{B+C}{\Gamma_g^2+p^2} + \frac{D+E}{\Gamma_g^2+4p^2} + \frac{F+G}{\Gamma_g^2+9p^2} \right) \cos \omega_o t + \left(\frac{A}{\Gamma g} + \frac{(C-B)}{\Gamma_g^2+p^2} + \frac{(E-D)}{\Gamma_g^2+4p^2} + \frac{(G-F)}{\Gamma_g^2+9p^2} \right) \sin \omega_o t \right\} , \tag{5}$$

where $\Gamma_\mu >> (g_F \omega_\ell - \omega_o)$, and where A, B, C, etc. are the expansion coefficients of $1/[\Gamma_g + i(\Delta n)p]$, and $p = (\delta^2 + b^2)^{1/2}$. Although the evaluation of the expansion coefficient is straightforward it is not reproduced here.

The experimental arrangement is identical to that of our previous experiment,[4] where we observed the transfer of only the longitudinal component of the alignment collisions. Figures 1 and 2 show the components of the absorbed light that are modulated at $\cos \omega_o t$ and $\sin \omega_o t$. The shapes of these curves are very similar to the B and C functions obtained

experimentally and theoretically by Dodd and Series.[7] Typical experimental conditions were: pressure of Xe = 5 microns, metastable state production electron current = 80 mA/cm^2, with 16 eV across the diode, and H_1 = about 8 mG. Excessive rf field, H_1, distorted the curve considerably. The experiment was performed at various frequencies ranging from 200 to 2000 Hz, which were crystal-oscillator-controlled.

The experimental result shows evidence of the transfer of the transverse component of the alignment through collisions. We believe that this experiment shows that the recently developed optical detection techniques together with the electron pumping technique (which has a very fast pumping speed compared with optical pumping) is a convenient method for studying the magnetic properties of the atoms inaccessible to either an optical or a magnetic induction method. Transfer of the transverse component of alignment should also occur in ionic ground state metastable-state collisions used in the experiment we performed previously.[8]

Footnotes and References

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[8] T. Hadeishi and C.-H. Liu, Phys. Rev. Letters 17, 513 (1966).

Figure Captions

Fig. 1. The antisymmetric ($\cos \omega_o t$) component of the modulated light-absorption signal as the magnetic field is varied. The applied frequency was 1000 Hz.

Fig. 2. The symmetric ($\sin \omega_o t$) component of the modulated light-absorption signal versus magnetic field. The applied frequency was 1000 Hz.

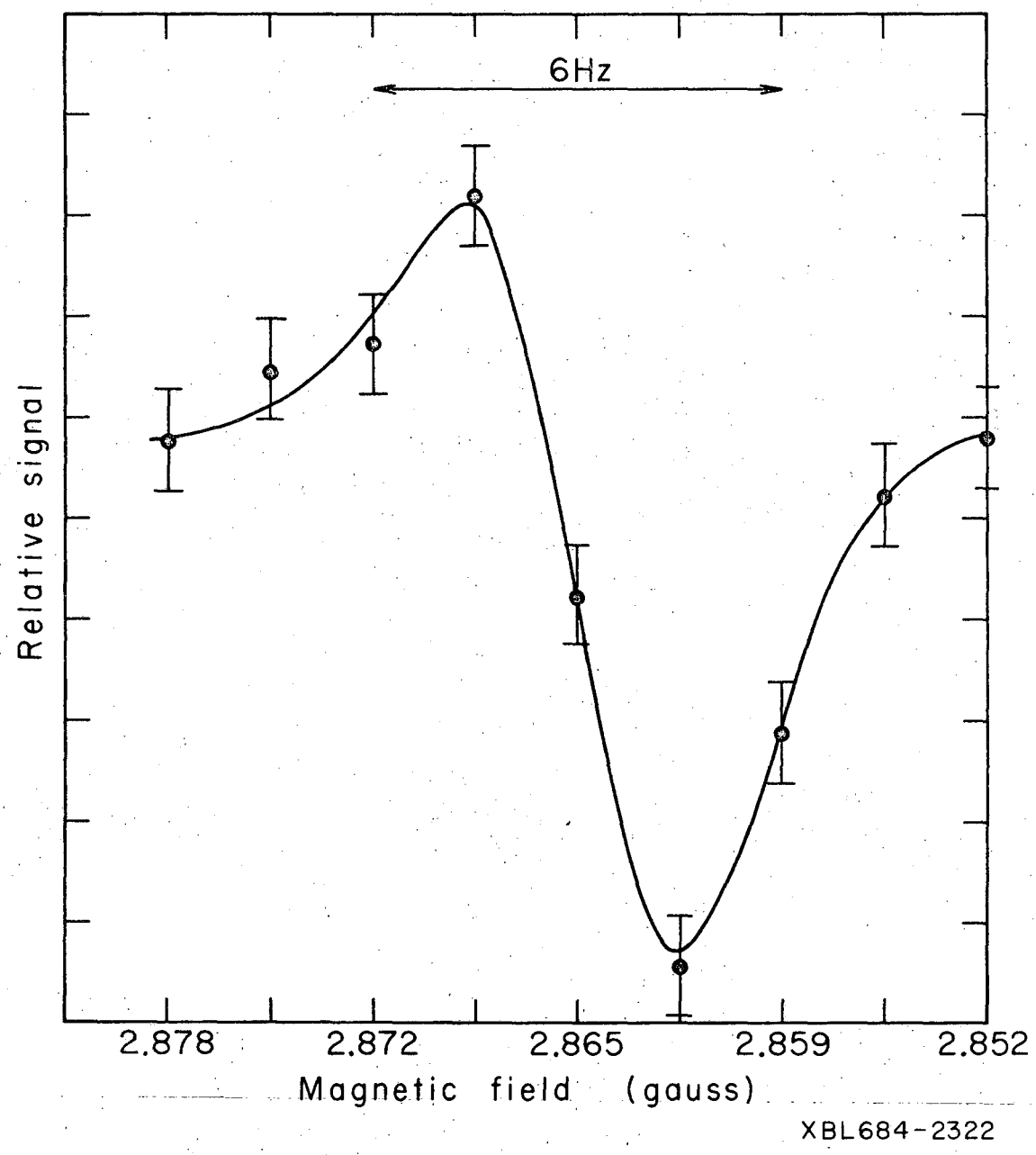
6Hz
Relative signal
2.878
2.872
2.865
2.859
2.852
Magnetic field (gauss)
XBL684-2322

Fig. 1

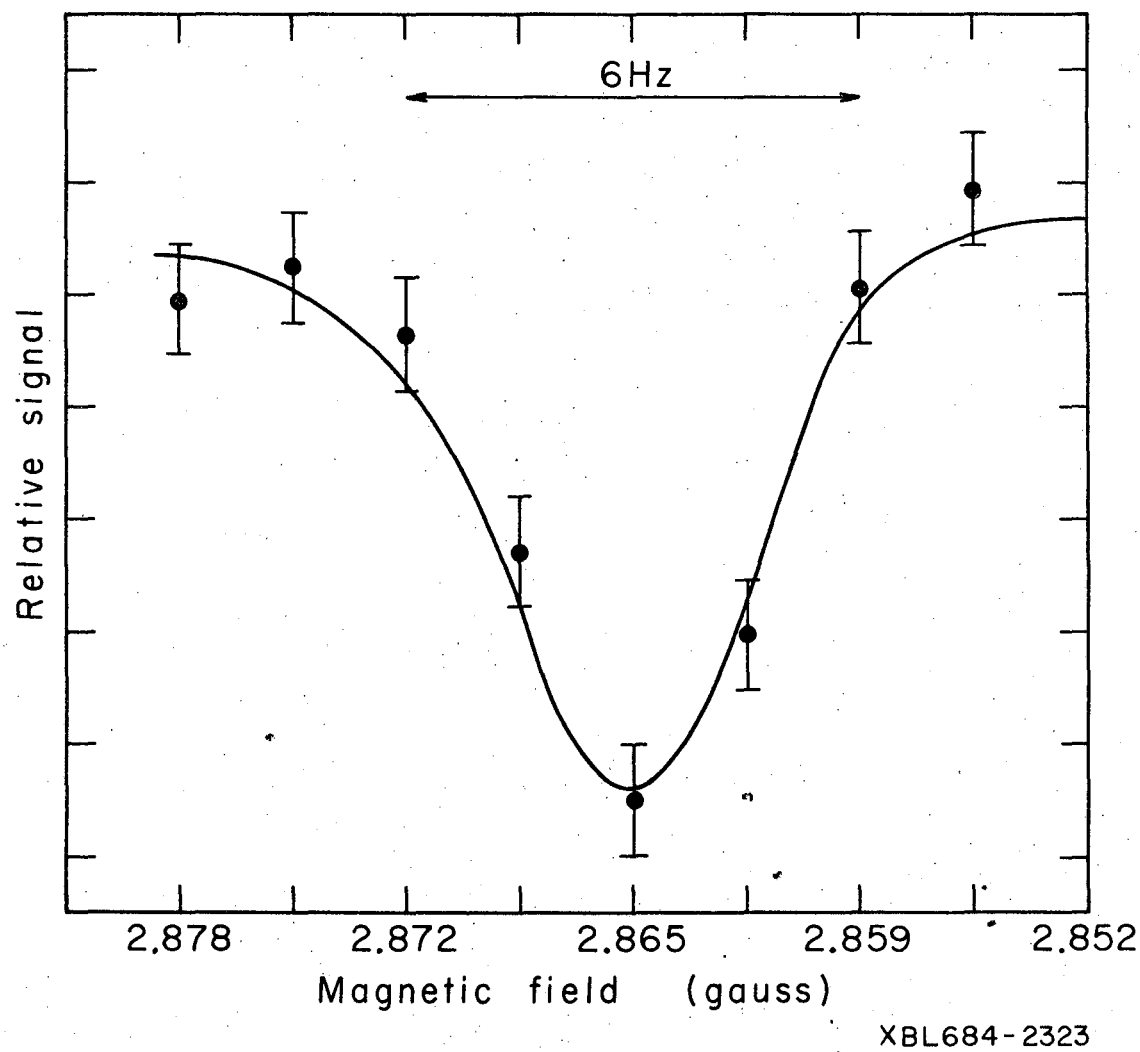
6Hz
Relative signal
2.878
2.872
2.865
2.859
2.852
Magnetic field (gauss)
XBL684-2323

Fig. 2

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