Nuclear Spin-Lattice Relaxation in the Presence of Magnetic-Field Gradients*

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(Received 9 April 1965)

Brownian motion of a spin in the presence of a magnetic-field gradient can significantly influence the spin-lattice relaxation time. A theory is developed to describe this effect, and experiments on gaseous He³ which confirm the theory in detail are reported. Practical situations where this mechanism can dominate, such as in the construction and use of polarized He³ targets in nuclear-scattering experiments, are discussed. In a homogeneous magnetic field, relaxation times of approximately 7 h at low pressures permit us to establish an upper limit of $7\times10^{-15}e$ cm for the electric-dipole moment of the He³ nucleus.

I. INTRODUCTION

A NUMBER of authors have considered the combined effects of translational diffusion and gradients in the applied magnetic field on the relaxation of nuclear spin systems. Such an effect, insofar as it influences the spin-spin relaxation time T_2, was analyzed by Hahn[1] and by Carr and Purcell[2] in conjunction with their definitive work on nuclear-resonance techniques involving free precession. Later, the phenomenological Bloch equations of nuclear magnetic resonance were generalized by Torrey[3] to include transfer of magnetization by diffusion.

Bloch was the first to point out that translational diffusion in the vicinity of strong irregular magnetic fields could contribute effectively to spin-lattice relaxation, and he proposed using paramagnetic powders as a catalyst for nuclear relaxation in monatomic gases.[4] This technique was successfully employed by Proctor and Yu to shorten the relaxation time of Xe¹²⁹, thus allowing measurement of the Xe magnetic moment by nuclear-magnetic-resonance techniques.[5]

Kleppner, Goldenberg, and Ramsey in connection with their work on the atomic-hydrogen maser derived approximate expressions for T_2 and T_1, the spin-lattice relaxation time, for the case of spins which are reflected from container walls in the presence of an inhomogeneous magnetic field.[6]

Quite a different situation from that considered by Kleppner *et al.* arises when the mean free path of the diffusing atoms is much smaller than container dimensions, and it is that case which we shall consider here. In their experiments on optical pumping and nuclear polarization in low-pressure He³ gas, Colegrove, Schearer and Walters observed that even very weak magnetic-field gradients completely control the He³ spin-lattice relaxation time.[7] Qualitatively, this decrease in T_1 was explained in terms of the fluctuating magnetic

field seen by a He³ atom as it undergoes Brownian motion in the presence of the field gradient.[8] Subsequent work in this area has been reported briefly by us[9] and by Gamblin and Carver.[10]

This paper presents a quantitative theory of spin-lattice relaxation caused by diffusion of spins in inhomogeneous magnetic fields, and describes experiments which verify in all respects the calculated dependence of T_1 on gas pressure, magnetic-field strength, and gradient strength.

The application of optical-pumping techniques to He³ gas at low pressures (several mm Hg) yields impressively large nuclear polarizations and has stimulated interest in the development of polarized He³ ion sources and target chambers suitable for use in nuclear scattering experiments.[11,12] The results reported here are of practical importance in the construction of such apparatus since the relaxation of the He³ nuclei by diffusion through gradients constitutes a "leak" of the angular momentum imparted to the He³ spin system by the circularly polarized pumping light. If the leak is sufficiently large, the polarization will be degraded. The effects of inhomogeneous magnetic fields are particularly apparent in the environment required for optical pumping. The relaxation formula derived in Sec. II provides the necessary guidelines for establishing toleration limits on magnetic-field gradients which might result from target or ion source construction materials and from inhomogeneity in the applied magnetic field.

II. THEORY

Brownian motion in the presence of magnetic field gradients causes the moving atoms to experience randomly fluctuating magnetic fields. Such fluctuating fields contribute to spin-lattice relaxation of the atomic nuclei. Using time-dependent perturbation theory, an

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[8] Equation 11 for T_1 in Ref. 7 is incorrect and should be replaced by Eq. 12 of this paper.
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[12] G. Roy, S. D. Baker, G. C. Phillips, and G. K. Walters, Bull. Am. Phys. Soc. 10, 51 (1965).

expression for the spin-lattice relaxation time T_1 can be derived for a system of spins I which do not interact with one another but do couple to random external field fluctuations. The applied magnetic field $\mathbf{H}$ is regarded as a superposition of a weak spatially varying field upon a much stronger homogeneous field $\mathbf{H}_0$. The perturbation Hamiltonian is

$$\mathcal{3C}(t) = -\gamma \hbar \mathbf{I} \cdot \mathbf{h}(t), \qquad (1)$$

where $\mathbf{h}(t)$ is the fluctuating field seen by a spin of gyromagnetic ratio γ as it executes Brownian motion in the presence of the gradient in the applied magnetic field $\mathbf{H}$. If $\mathbf{h}(t)$ is a random stationary function of time, the standard treatment for relaxation problems of this kind may be followed,[13] yielding

$$1/T_1 = (2/\hbar^2) P(\omega_0), \qquad (2)$$

where $\omega_0 = \gamma H_0$ is the average magnetic resonance frequency, and $P(\omega_0)$ is the spectral density function of the matrix elements of the perturbation Hamiltonian, given by

$$P(\omega) = \int_{-\infty}^{+\infty} \langle A_{km}(t) A_{km}{}^*(t+\tau) \rangle_{\mathrm{av}} e^{-i\omega\tau} d\tau \qquad (3)$$

with

$$A_{km}(t) = -\gamma \hbar \sum_{q=x,y} h_q(t)(k|I_q|m); \qquad (4)$$

$h_x(t)$ and $h_y(t)$ are the x and y components of the field fluctuation $h(t)$. If we consider a spin-$\frac{1}{2}$ system and assume that the components $h_q(t)$ of the fluctuating field are statistically independent, the time-independent matrix elements $(\pm\frac{1}{2}|I_q|\mp\frac{1}{2})$ can be evaluated and the T_1 expression becomes

$$1/T_1 = \tfrac{1}{2}\gamma^2 J(\omega_0), \qquad (5)$$

where

$$J(\omega_0) = \int_{-\infty}^{+\infty} \langle [h_x(t)+h_y(t)]$$
$$\times [h_x(t+\tau)+h_y(t+\tau)] \rangle_{\mathrm{av}}\, e^{-i\omega\tau} d\tau. \qquad (6)$$

Because $h_x(t)$ and $h_y(t)$ vary independently, the cross terms in $J(\omega_0)$ have zero average, and

$$\frac{1}{T_1} = \frac{\gamma^2}{2} \int_{-\infty}^{+\infty} \{\langle h_x(t) h_x(t+\tau)\rangle_{\mathrm{av}}$$
$$+ \langle h_y(t) h_y(t+\tau)\rangle_{\mathrm{av}}\} e^{-i\omega\tau} d\tau. \qquad (7)$$

Thus, the problem reduces to finding the spectral densities of the autocorrelation functions $g_q(\tau) = \langle h_q(t) h_q(t+\tau)\rangle_{\mathrm{av}}$.

In the case considered here, the field fluctuations occur as a result of the Brownian motion of the spins in the presence of a magnetic-field gradient. For simplicity we consider a field gradient which is constant and

[13] C. P. Slichter, *Principles of Magnetic Resonance* (Harper and Row, New York, 1963), Chap. 5.

FIG. 1. The displacement of the atom from P to P' due to its velocity U_y generates a rotation of the magnetic field as seen by the atom at the rate $d\theta/dt = \omega_R$. For small gradients, $\theta \approx H_y/H_0$ and $\omega_R = (1/H_0)(\partial H_y/\partial y) \times U_y(t)$. Collisions occurring during the motion of the atom cause $U_y(t)$ and consequently ω_R to fluctuate in a random manner.
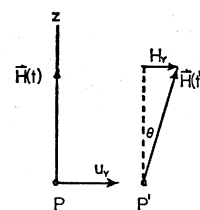
axially symmetric with respect to the sample and with the variation of H over the sample being small compared to H_0.[14]

As discussed later, our experimental conditions easily met these requirements. Under such circumstances, the autocorrelation functions $g_x(t)$ and $g_y(t)$ are equal, and their spectral density functions may be evaluated in either of the following equivalent ways. (While this manuscript was in preparation, the writers learned that Gamblin and Carver, by means of a classical random-walk calculation have independently obtained an expression for T_1 which is in agreement with the result derived below to within a numerical factor of order unity.[10])

A. Rotating-Reference-Frame Derivation

Because of the spread in field directions over the sample volume, there is no uniquely defined axis of spin quantization in the laboratory reference frame. We consider a spin initially at an arbitrary point in the sample, and choose the z axis at that point to lie along the local magnetic-field direction. Let the y axis be chosen so that the velocity vector of the atom lies in the y-z plane. As the spin moves away from its initial position with velocity $\mathbf{U}$ it sees the field direction rotating about the x axis with an angular velocity (see Fig. 1)

$$\omega_R = (1/H)(\partial H_y/\partial y) U_y. \qquad (8)$$

We wish to view the problem in a reference frame in which the z axis always coincides with the local magnetic-field direction as seen at any instant by the moving spin; that reference frame is simply one which rotates about the x axis with angular velocity ω_R. In this rotating reference frame, an effective field

$$H_{\mathrm{eff}} = \omega_R/\gamma = (1/\gamma H)(\partial H_y/\partial y) U_y \qquad (9)$$

appears in the x direction.

Now consider the effect of an atomic collision which alters the particle velocity. The spin starts moving in a new direction, and to keep the z axis coincident with the local magnetic-field direction we must abandon our original rotating reference frame and transform to a new frame which rotates about an axis in the transverse plane which is perpendicular to the new transverse

[14] This statement is unnecessarily restrictive since the statistical nature of the process is established after a few collisions. It can be shown that the appropriate restiction is actually to limit to small values the variation in the magnetic field over the distance the atom diffuses in several mean free paths.

velocity. H_{eff} then points in a new direction in the x-y plane. Subsequent collisions require similar coordinate transformations. Thus, in a reference frame where the z axis is always along the local magnetic field direction, motion through gradients in H causes a magnetic field H_{eff} to appear in the x-y plane and collisions cause H_{eff} to fluctuate in both direction and magnitude. This fluctuating transverse effective field can induce magnetic dipole transitions, and it is the autocorrelation function for $H_{\text{eff}}(t)$ that we require in Eq. (7) for the relaxation time T_1. The autocorrelation function is just

$$\langle h_x(t)h_x(t+\tau)\rangle_{\text{av}}=[(1/\gamma H_0)(\partial H_y/\partial y)]^2$$
$$\times\langle U_y(t)U_y(t+\tau)\rangle_{\text{av}}. \quad (10)$$

$U_y(t)$ is the stochastic variable representing the velocity of a particle undergoing Brownian motion and its autocorrelation function is well known to be[15]

$$\langle U_y(t)U_y(t+\tau)\rangle_{\text{av}}=\langle U_y^2\rangle e^{-|\tau|/\tau_c}, \quad (11)$$

where τ_c is the mean time between atomic collisions.[16] Substitution of Eqs. (10) and (11) into the T_1 expression and evaluation of the integral yields

$$1/T_1=(2/3H_0^2)(\partial H_y/\partial y)^2\langle U^2\rangle_{\text{av}}[\tau_c/(1+\omega_0^2\tau_c^2)], \quad (12)$$

where we have used for the mean-squared velocity

$$\langle U^2\rangle_{\text{av}}=3\langle U_x^2\rangle_{\text{av}}=3\langle U_y^2\rangle_{\text{av}}$$

and, by symmetry,

$$(\partial H_y/\partial y)=(\partial H_x/\partial x).$$

B. Laboratory-Reference-Frame Derivation

Alternatively, the T_1 expression may be derived using the laboratory reference system. Since there is no unique axis of spin quantization, let us choose an origin of coordinates at an arbitrary point in the sample with the z axis parallel to H_0. Because of the gradient in H, a spin moving about in the sample container sees variable x and y components of field relative to the fixed coordinate axes just defined. In terms of its position coordinates, the magnetic field seen by a spin is

$$\mathbf{H}=-\mathbf{i}\mathcal{G}x-\mathbf{j}\mathcal{G}y+\mathbf{k}(2\mathcal{G}z+H_0), \quad (13)$$

where $\mathcal{G}=\partial H_x/\partial x=\partial H_y/\partial y$ gives the magnitude of the constant field gradient. Thus

$$h_x(t)=-\mathcal{G}x(t) \quad \text{and} \quad h_y(t)=-\mathcal{G}y(t),$$

and

$$J_x(\omega)=\mathcal{G}^2\int_{-\infty}^{+\infty}\langle x(t)x(t+\tau)\rangle_{\text{av}}\,e^{-i\omega\tau}\,d\tau.$$

However, the displacement of a free particle undergoing Brownian motion does not represent a random stationary process and the quantity $\langle x(t)x(t+\tau)\rangle_{\text{av}}$ does not exist. Fortunately, we do not require $\langle x(t)x(t+\tau)\rangle_{\text{av}}$; we require only its Fourier transform. The important point is that even though $x(t)$ does not represent a random stationary process, it does represent a random process with stationary increments. [Physically, we do not care about the position $x(t)$ of the spin; rather, we are concerned with the fluctuations in $x(t)$, which are related to the incremental motion.] Mathematically, one can evaluate the spectral density function by introducing an elastic restoring force which acts on the particle; $x(t)$ then describes the Brownian motion of a simple harmonic oscillator, a process which is random and stationary. The autocorrelation function is well-known and its Fourier transform is easily evaluated.[15] Upon taking the Fourier transform, one lets the force constant go to zero and the result is the desired spectral density function for a free particle,[17]

$$\int_{-\infty}^{\infty}\langle x(t)x(t+\tau)\rangle_{\text{av}}e^{-i\omega\tau}d\tau=2\langle U_x^2\rangle_{\text{av}}\frac{\tau_c}{\omega^2(1+\omega^2\tau_c^2)}, \quad (14)$$

where $\langle U_x^2\rangle_{\text{av}}$ is the mean-squared x component of particle velocity, and τ_c is the mean time between atomic collisions. Using $\langle U_x^2\rangle=\frac{1}{3}\langle U^2\rangle$ and substituting into Eq. (7), we obtain

$$\frac{1}{T_1}=\frac{2}{3}\gamma^2\mathcal{G}^2\langle U^2\rangle\frac{\tau_c}{\omega_0^2(\omega_0^2\tau_c^2+1)}$$

which is identical to the result obtained in part A.

III. EXPERIMENT

The theory developed in Sec. II was verified experimentally by spin-lattice relaxation time studies of gaseous He^3 samples subjected to known magnetic field gradients. To test the predicted dependence of T_1 on τ_c and H_0, measurements were made over a range of He^3 gas pressures from 0.2 to 10 mm Hg, and for magnetic-field strengths ranging from 1 to 240 G. The He^3 samples were contained in 60-cc Pyrex spheres.

A. Techniques

Relaxation times were measured by monitoring the decay of He^3 spin magnetization from an initially highly polarized state. Optical pumping methods were used to produce the spin polarization. A detailed description of optical pumping in He^3 may be found elsewhere.[7] Briefly, the polarization technique is as follows: A weak electrical discharge in the He^3 sample cell excites a small fraction of the He^3 atoms to the 2^3S_1 metastable state. Circularly polarized 2^3S_1-2^3P_0 resonance radiation, incident along the magnetic-field direction, induces

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[15] M. C. Wang and G. E. Uhlenbeck, Rev. Mod. Phys. **17**, 323 (1945).

[16] Actually the autocorrelation function desired is $\langle U_y(t)\times U_{y'}(t+\tau)\rangle_{\text{av}}$ where the y and y' axes differ as a result of the rotating-coordinate transformation employed. However, we have already assumed that the variation of H over a collision mean free path is very small compared to H; hence $\omega_R\tau_c\ll 1$ and the y and y' axes are essentially coincident for time intervals of several τ_c during which the autocorrelation function has nonnegligible value. Thus, we are justified in replacing $\langle U_y(t)U_{y'}(t+\tau)\rangle_{\text{av}}$ by $\langle U_y(t)U_y(t+\tau)\rangle_{\text{av}}$.

[17] The justification for this step is outlined in A. M. Yaglom, *An Introduction to the Theory of Station Random Functions* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1962), p. 92ff.

spin polarization of the metastables. This polarization is in turn transferred by collisions involving electron exchange to the more numerous ground state atoms. In steady state the ground-state atoms attain the same polarization as the metastables. For the experiments reported here, He^3 polarizations between 5 and 10% were typical.

To measure T_1, the sample was first polarized by the optical pumping technique. Then the electrical discharge was turned off, thus providing the desired highly polarized state of the unexcited He^3 gas. The sample magnetization then decays with time constant T_1 to its thermal equilibrium value. In the absence of magnetic-field gradients the decay times were always greater than 1 h, spin relaxation in that case resulting from interactions of the He^3 nuclei with the container walls. The gradient-dominated relaxation times were always much smaller, so that wall relaxation effects could be safely neglected.

The magnetization measurements were made in a 220-G magnetic field and in the following sequence. Immediately after turning off the electrical discharge, the nuclear-magnetic-resonance signal is observed by conventional slow passage NMR techniques at 714 kcps. The strength of the NMR signal is proportional to the initial magnetization M_0. The applied magnetic field is then changed to the value H_0 at which the gradient-dominated relaxation time is to be measured, and the known gradient is applied for a time Δt. After the gradient is turned off, the field is returned to 220 G, and the residual magnetization M is measuerd. The relative strengths of the intial and residual NMR signals, along with Δt, allow calculation of T_1, assuming an exponential decay. The entire sequence was always completed in a time much less than the relaxation time of the He^3 in the absence of gradients.

The external field for the technique described above was provided by an end-corrected solenoid with a homogeneity over the sample of about 1 part in 10^5. The gradient was provided by a large set of Helmholtz coils connected in opposition; this provided an easily calculable constant gradient over the sample at the center of the coils, so that the total field seen by the sample was given in Eq. (13).

An alternative optical method for measuring T_1 was also used. The measurement sequence was the same as that previously described with the exception that the sample polarization was determined optically instead of by NMR. Optical determination of the polarization is possible because the percentage of the 2^3S_1-2^3P_0 pumping light that is absorbed by the sample depends on the degree of polarization of the sample. The physical basis for this dependence and the experimental techniques for measuring the polarization optically are fully discussed in Ref. (7).

The optical method has the disadvantage that the electrical discharge must be turned on to populate the 2^3S_1 state prior to making the absorption measurement.
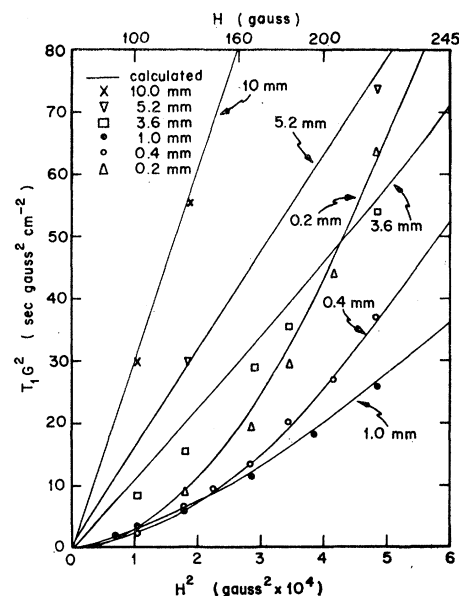

FIG. 2. Relaxation by gradients as a function of applied field at various pressures. The solid lines in each case are calculated from Eq. (12). The vertical scale is plotted in units of $T_1\mathcal{G}^2$ where $\mathcal{G}$ is the gradient $\partial H_x/\partial x$. The gradient was chosen to be either 1.6 or 0.8 G cm^{-1} to keep T_1 within convenient limits. The value of τ_c is obtained from the minima of the curves shown in Fig. 3. The value of τ_c can also be obtained by considering the crossovers of the various curves.

For the residual polarization determination after application of the magnetic gradient, the discharge must be re-ignited and the measurement made in a time short enough so that there is no appreciable change in the polarization due to repumping by the measuring light. Typically, the optical pumping time exceeds 30 sec, and the polarization measurement can be made in about 1 sec. An advantage of the optical technique over the NMR method is the greater accuracy obtainable because of better signal-to-noise ratio.

B. Results

He^3 relaxation times for gradients up to 1 G/cm were measured over a range of pressures from 0.2 to 10 mm Hg and in fields from 1 to 240 G. These ranges are sufficiently broad to test the theory over the entire range from $\omega_0\tau_c \ll 1$ to $\omega_0\tau_c \gg 1$. The experimental results are summarized in Figs. 2 and 3. The solid lines are derived from Eq. (12). In calculating the theoretical curves, measured values of H_0 were used and the gradient was calculated from the coil geometry and current. The mean-squared velocity was taken as $\langle U^2 \rangle = (3kT)/m$. All measurements were made at 300°K. The only remaining variable on the right hand side of Eq. (12) is τ_c, the mean time between collisions, which varies inversely with the pressure. Pressures were measured at the time of sample preparation using a high-pressure thermocouple gauge previously calibrated against a precision McLeod gauge. The relation between τ_c and
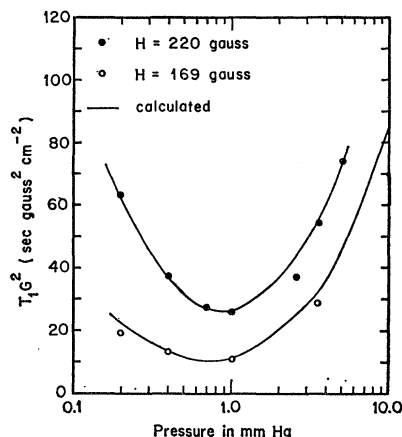
FIG. 3. Relaxation by gradients as a function of pressure. The solid curves are calculated from Eq. (12). A best fit to the experimental points shown gives $\tau_c = 2.2 \times 10^{-7}$ sec at 1 mm Hg assuming T_1 is a minimum at $\omega_0 \tau_c = 1$.

reciprocal pressure could be calculated from the known helium-helium elastic-collision cross section derived from gas-kinetic interpretation of viscosity and diffusion data; however, we instead determined the proportionality constant from the data of Fig. 3 by requiring that the minimum in the theoretical curve of T_1 versus pressure at $\omega_0 \tau_c = 1$ coincide with the T_1 minimum found experimentally. This procedure gives $\tau_c = (2.2 \pm 0.2) \times 10^{-7} p^{-1}$ sec at 300°K, where p is in mm Hg. The elastic-collision cross section is then found from $\sigma = (\tau_c v \; n)^{-1}$ where v is the rms atomic velocity and n the density of the He^3 in atoms/cc. In view of the excellent agreement between theory and experiment, it is reasonable to regard this work as providing a new independent determination of τ_c. It is in good agreement with the values calculated from viscosity and diffusion data.

An interesting feature of this relaxation mechanism is the change in T_1 dependence from $T_1 \sim p H_0^{+2}$ when $\gamma H_0 \tau_c \ll 1$ to $T_1 \sim p^{-1} H_0^{+4}$ for $\gamma H_0 \tau_c \gg 1$. This accounts for the crossovers that occur between curves representing different pressures in Fig. 1.

As an example of a typical experimental measurement, a gradient of 1.5 G cm^{-1} gave a relaxation time of 20 sec for He^3 gas at 0.4 mm pressure in an H_0 field of 240 G. At 110 G, a gradient of 0.43 G cm^{-1} produces the same T_1.

This relaxation mechanism becomes extremely important in low-pressure gases in weak magnetic fields where even the gradients due to nearby test equipment may be sufficient to drastically affect T_1.

CONCLUSIONS

The fluctuating magnetic field seen by a spin executing Brownian motion in the presence of a magnetic field gradient contributes to spin-lattice relaxation. The theory developed in Sec II provides a quantitative relationship between T_1, gas pressure, magnetic-field strength, gradient strength, and thermal velocity. The experimental results reported in Sec. III verify the theory in every respect over a wide range of all variables.

Under normal circumstances involving NMR studies of high-pressure gases and liquids using normal laboratory magnetic fields, gradient relaxation is usually negligible. However, for gases at low pressures and in weak magnetic fields, gradient relaxation is dominant unless one exercises extreme care in providing a highly homogeneous magnetic field over the sample and keeps all ferromagnetic materials out of the general vicinity of the sample. Experiments involving optical pumping of He^3 gas are generally done under such low pressure and weak field conditions, and the He^3 nuclear polarization attainable by optical pumping is seriously degraded if gradients are not assiduously avoided.

In particular, one must avoid even slightly ferromagnetic materials in the construction of He^3 optical pumping samples. This is especially troublesome in the development of polarized He^3 ion sources,[18] and in the design of polarized He^3 target cells[11] suitable for use in nuclear scattering experiments where one is often faced with the need for thin metallic foils, electrical feedthroughs, gas valving, and particle counters within the target chamber.

Similar care must be exercised in operation of the optically-pumped He^3 maser, first reported by Robinson and Myint.[19]

Purcell has shown that if a nucleus possesses an electric-dipole moment that relaxation can occur via the interaction of the electric dipole with the fluctuating electric field at the nucleus during collisions.[20] The relaxation time in this case is given by

$$\frac{1}{T_1} = \frac{8\mu_e^2 mkT \omega_0^2 \tau_c}{\hbar^2 Z^2 e^2 (1 + \omega_0^2 \tau_c^2)} \tag{15}$$

In the absence of magnetic-field gradients, the relaxation time of the He^3 nuclear spins in a carefully prepared sample at 1 mm Hg and 240 G ($\omega_0 \tau_c = 1$) is greater than 6.7 h. Using Eq. (15) we can set a new upper limit on the size of the electric-dipole moment of the He^3 nucleus $\mu_e < 7 \times 10^{-15} e$ cm.

ACKNOWLEDGMENTS

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