

Nuclear Magnetic Resonance of Xe^{129} in Natural Xenon*†

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The spin-lattice relaxation time T_1 of Xe^{129} has been measured as a function of temperature in the liquid and as a function of pressure in the gas. A strong shift ΔH in the external field required for resonance at constant frequency but varying sample density ρ has been discovered. As the sample density increases in the region above 48 atm, ΔH decreases linearly at the rate of 3.45 milligauss per amagat (density at standard conditions) in a field of 8060 gauss. In the liquid the temperature dependence of the product of T_1 and ρ can be described by an activation energy of 0.7 ± 0.1 kcal/mole. In the gas at room temperature between 48 and 73 atm, T_1 varies as $\rho^{-2.1 \pm 0.4}$. The largest value of T_1 observed was 2600 ± 600 sec for a gas sample at 48 atm, and the shortest value

was 57 ± 2 sec in the liquid at -101°C . The experimental values of T_1 have been compared with theoretical predictions in two limiting cases, the rare gas and the dense liquid. In both cases the experimental values, although larger than previously reported values, are still two to three orders of magnitude smaller than predicted from an intermolecular nuclear magnetic dipole interaction. The relaxation time was found to be independent of field. Implications of our data for determining the relaxation mechanism are discussed. It is suggested that the relaxation may be caused by a fluctuating magnetic field at the nucleus resulting from the motion of nonsymmetrical electronic charge distributions during collisions.

I. INTRODUCTION

STUDIES of nuclear spin relaxation times can reveal important information concerning the microscopic properties of liquids and gases.¹⁻⁴ Liquid and gaseous xenon samples are particularly interesting.

Xenon is expected to be an excellent system for investigation because of the simplicity of both its general structure and its anticipated spin relaxation mechanism. The xenon molecules are monatomic and spherically symmetric. They are sufficiently massive that the intermolecular collisions do not exhibit major quantum effects. As a result xenon represents the simplest type of classical fluid. The nuclear spin relaxation involves several simplifications. Two xenon isotopes found abundantly in natural samples have nuclear magnetic dipole moments. They are Xe^{129} (26.2%) and Xe^{131} (21.2%). Of these, Xe^{129} has spin $\frac{1}{2}$ and no electric quadrupole moment. The Xe^{129} spin relaxation is, therefore, free from complications due to electric quadrupole effects. Also, because xenon is monatomic, the relaxation is free from complications due to intramolecular nuclear dipole fields with their time variation determined by rotational motion. Only the intermolecular nuclear dipole fields with their time variation determined predominantly by translational diffusion are expected to provide a relaxation mechanism. Torrey^{3,4} has developed a detailed theory for relaxation by this mechanism. Although xenon is a simple system,

it is difficult to work with experimentally because the nuclear signals are weak and the relaxation times long.

There is a second reason for immediate interest in xenon. Despite the fact that the theoretical predictions of nuclear spin relaxation times for Xe^{129} in pure xenon samples are expected to be quite straightforward, Staub's^{5,6} early measurements on gaseous xenon revealed Xe^{129} relaxation times differing from the predicted values by four orders of magnitude. This large discrepancy does not exist for Xe^{131} relaxation which can be accounted for in terms of an electric quadrupole effect during collisions.^{7,8} It is possible that Xe^{129} possesses an unexpected relaxation mechanism quite different from and more effective than the direct nuclear dipole mechanism. On the other hand, since the predicted relaxation times in the rare gas are quite long, small amounts of an impurity, such as oxygen, either in the fluid or on the container walls might act as a dominant source of relaxation.

Since an understanding of the relaxation mechanism is a prerequisite to further important nuclear resonance studies of xenon, we initiated the present work in an effort to learn more about Xe^{129} relaxation under a variety of conditions.

II. THE XENON SAMPLES

Six natural xenon samples having room temperature pressures from 30 to 109 atm were prepared for this work. Xenon gas was slowly admitted to and frozen in small Pyrex bulbs having a 0.4-cm inner diameter and a 1-cm length. Each bulb was at one end of a long capillary. After the desired amount of gas had been admitted and frozen, the other end of the capillary about 20 cm from the bulb was sealed. The volume of a bulb

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‡ Now at the National Bureau of Standards, Washington, D. C.

¹ N. Bloembergen, *Nuclear Magnetic Relaxation* (Martinus Nijhoff, The Hague, 1948).

² N. Bloembergen, E. M. Purcell, and R. V. Pound, *Phys. Rev.* **73**, 679 (1948).

³ H. C. Torrey, *Phys. Rev.* **92**, 962 (1953).

⁴ H. C. Torrey, *Suppl. Nuovo cimento* **9**, 95 (1958).

⁵ E. Brun, J. Oeser, H. H. Staub, and C. G. Telschow, *Phys. Rev.* **93**, 904 (1954).

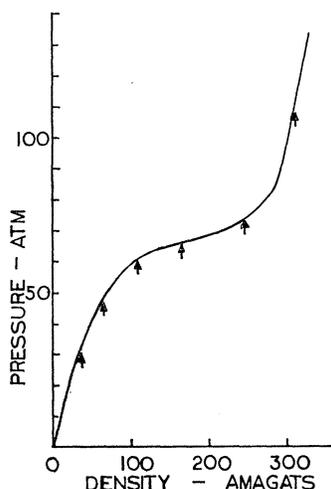
⁶ E. Brun, J. Oeser, H. H. Staub, and C. G. Telschow, *Helv. Phys. Acta* **27**, 173 (1954).

⁷ F. J. Adrian, thesis submitted to the faculty of Cornell University, 1955 (unpublished).

⁸ H. Staub, *Helv. Phys. Acta* **29**, 246 (1956).

and capillary was measured to within 0.5%. The mass of the gas admitted was measured by two methods; by weighing the samples before and after the filling procedure and by measuring the volume of gas at room temperature and atmospheric pressure before it was admitted to the bulb. These two measurements agreed to within 1%, but the weighing provided the more accurate means for determining the density of the sample. Figure 1 is a plot of the pressure of xenon as a function of density at 25°C. The density is given in the relative unit of an amagat which corresponds to the gas at 0°C and one atmosphere of pressure. For xenon one amagat unit of density is 4.49×10^{-5} mole/cc. This is within about 1% of the ideal gas value. The mass density at these standard conditions is 0.00589 g/cc. The small arrows in the Fig. 1 indicate the densities and pressures of our six samples. Precise data on the six samples are given in Table I.

FIG. 1. Xenon pressure as a function of density at 25°C from A. Michels, T. Wassenaar, and P. Louwerse, *Physica* 20, 99 (1954), and J. A. Beattie, R. J. Brierley, and J. S. Brierley, *J. Chem. Phys.* 19, 1219 (1951). The arrows indicate points corresponding to our samples.



We have observed nuclear magnetic resonance in liquid xenon as a function of temperature by lowering the temperature of the above samples below the critical temperature of 17°C. The critical pressure is 58 atm. As the temperature decreases below room temperature, the sample remains at its uniform density until it condenses into two phases. The condensation temperature for a particular density is given by the coexistence curve of Fig. 2. For still lower temperatures, the density of the liquid phase is given by the high-density side of the coexistence curve. The ratio of the capillary to bulb volumes for our sealed samples is such that below the condensation point the bulb is entirely filled with the liquid phase. Since the nuclear resonance coil surrounds only the bulb, the observed relaxation times are those of the liquid phase.

In order to obtain as large a liquid range as possible, one uses a sample whose density is near to the critical density 1.11 g/cc. In the present work we have used our 1.44 g/cc sample. From Fig. 2 it is seen that the liquid

TABLE I. Density dependence for Xe¹²⁹ at 25°C and 8060 gauss.

Sample	Pressure (atm)	Density (g/cc)	Density (amagats)	T_1 (sec)	$\Delta H = H - H_r$ (gauss)
A	30	0.193	33	...	0.96 ± 0.06
B	48	0.386	66	2600 ± 600	0.90 ± 0.06
C	61	0.648	111	900 ± 180	0.73 ± 0.06
D	66	0.959	167	420 ± 180	0.53 ± 0.03
E	73	1.44	246	160 ± 30	0.30 ± 0.03
F	109	1.81	310	...	0.06 ± 0.03

range for this sample begins at approximately 15°C and continues down to the triple point at -112°C. If one works with a sample at atmospheric pressure, the liquid range extends only from -107°C to -112°C. It is interesting to note that xenon expands on freezing. The density of the liquid at the triple point is 3.05 g/cc and the density of the solid is 2.7 g/cc.

The mass spectrometric analysis of our assayed reagent-grade xenon obtained from the Matheson Company indicated that the number of impurity oxygen molecules was less than one part in 10^5 . In order to remove oxygen from the bulb volume and surfaces, the bulb was pumped down to 30 microns pressure and then flushed twice with helium gas. The estimated maximum amount of oxygen which could leak into the system while the pump was off during the sealing process was also the order of one part in 10^5 . However, the amount of oxygen finally remaining in each sample is still less. The temperature used to freeze out the xenon gas was sufficiently high that oxygen did not freeze out. Just before the seal was made, the sample was again pumped down to 0.03 mm. Depending on precisely how much oxygen was trapped in the frozen xenon, the oxygen impurity in our samples may be as low as one part in 10^8 ; one part in 10^6 is a liberal upper limit.

III. METHODS OF MEASUREMENT

We have used two methods to measure the Xe¹²⁹ spin-lattice relaxation times. In the measurements of

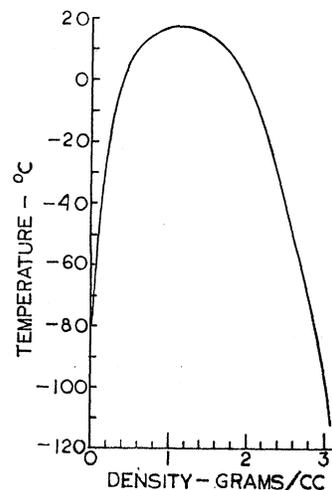


FIG. 2. Liquid-vapor coexistence curve for xenon from data in *International Critical Tables*.

the gas relaxation times as a function of pressure, the free-precession null technique was used.⁹ In this method the sample is first allowed to come to equilibrium with its magnetic polarization parallel to a strong external static magnetic field H_0 . Then the equilibrium polarization is inverted 180° by a strong short rf pulse. At the time $T_1 \ln 2$ the polarization will be zero as it returns to its equilibrium value. By measuring this null time one obtains T_1 .

To determine the null time, a 90° pulse is applied at varying times following the initial 180° pulse. The 90° pulse rotates the polarization into the plane perpendicular to the external field. This polarization then precesses freely about the external field. From the magnitudes of the induced nuclear rf signals, the time of the null can be identified.

This free-precession null method was also used to measure gas relaxation times at room temperature as a function of magnetic field. The samples were allowed to relax in the variable field of a second magnet. The polarization was then measured by applying the necessary pulses and observing the free-precession signal in the first magnet which had a fixed resonant field corresponding to the frequency of the equipment.

The above null method is destructive. Once the polarization is in the plane perpendicular to the external field, the polarization normally decays in a time much shorter than any of the relaxations times of the gas or liquid samples. This decay is related to the inhomogeneity of the external field H_0 .

After each measurement it is necessary to wait approximately six relaxation times before repeating the procedure. Otherwise, the sample will not return to its equilibrium value before the application of the new 180° pulse. For our xenon samples with relaxation times between one minute and one hour, this is an extremely time consuming and experimentally difficult procedure. We have, therefore, modified the above null method with a nondestructive measuring technique. At a time τ following each 90° pulse we apply a 180° pulse using a rf signal which is phase coherent with that used for the 90° pulse. Our value of τ is much shorter than the relaxation times being measured but equal to or slightly larger than the decay time associated with the inhomogeneity of the magnet. A spin echo forms.⁹ Just as the echo attains its maximum amplitude at a time 2τ following the first 90° pulse, we apply a second 90° phase coherent rf pulse. This rotates the magnetic polarization vector back to its original position parallel to the external field. As long as the pulse adjustments are accurate and the time 2τ is small compared to the relaxation times of the sample, the value of the polarization at the end of the measuring procedure is equal to that at the beginning. This procedure is nondestructive. In theory, one should be able to measure within one relaxation event ten or twenty points on the relaxation

curve. In practice such a procedure involves an accumulation of the errors related to imperfect pulses. One should be able to eliminate this accumulation by omitting the 90° pulse at 2τ and using a 180° pulse at 3τ and shifting by 180° the phase of the rf signal in both this pulse and a last 90° pulse applied at 4τ .

A second method for measuring spin-lattice relaxation times was used for our liquid samples. This involves a modification¹⁰ of the steady-state free precession technique.¹¹ Two advantages related to improvements in the nuclear signal strength are particularly important for the weak xenon signals. First, as in transient free-precession techniques, the effect of the inhomogeneity of the external magnet on signal strength can be eliminated. Second, as in steady-state forced precession techniques, the steady-state signal can be received through a narrow bandwidth with a resulting improvement in the signal-to-noise ratio. In the present work we have used the transient from zero polarization to a finite steady-state free precession polarization. It

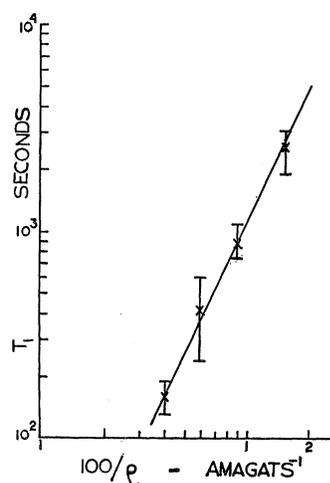


Fig. 3. Log-log plot of Xe^{139} relaxation time T_1 in the gas as a function of reciprocal density. The magnetic field is 8060 gauss and the temperature 25°C .

can be demonstrated theoretically that this transient growth is exponential with the time constant T_1 .¹⁰ In some cases we observe the transient when an unpolarized sample is initially placed in the magnet; in other cases a polarized sample already in the magnet is saturated with rf pulses before starting the observation.

This method represents a substantial improvement since the bandwidth is determined by the xenon spin-lattice relaxation time (~ 10 – $10\,000$ sec) rather than, as is the case for the null method, by the decay time due to the inhomogeneity of the magnetic field (~ 0.05 sec). We have not as yet been able to exploit this method fully for relaxation times over 200 sec.

In the present work we have made one exploratory measurement of the self-diffusion coefficient of liquid xenon. The nuclear resonance method described by Carr and Purcell⁹ was used.

¹⁰ R. L. Streever, thesis submitted to the faculty of Rutgers, The State University, 1959 (unpublished).

¹¹ H. Y. Carr, Phys. Rev. **112**, 1693 (1958).

⁹ H. Y. Carr and E. M. Purcell, Phys. Rev. **94**, 630 (1954).

As reported in the next section, resonance field shifts as a function of density have been observed. These shifts were measured by obtaining audio beats between the rf nuclear signals from the samples and a continuous rf leakage signal from our signal generator. A set of Helmholtz coils provides a small additional contribution to H_0 . The current control for these coils is a sensitive means of adjusting the precessional frequency of the nuclei. This current was initially adjusted to give a beat frequency of zero between the leakage signal and the nuclear signal from a reference sample. The shifts relative to the reference sample were determined from the change in Helmholtz coil current required to bring the beat frequency to zero for the other samples which were substituted for the reference sample.

IV. RESULTS

The Xe¹²⁹ relaxation times for the gas samples at room temperature in a 8060-gauss field are given in Table I and plotted in Fig. 3. An estimated maximum error is given with each value. In Fig. 3, T_1 as a function of $1/\rho$ is plotted on a log-log scale. The density ρ is

TABLE II. Temperature dependence for Sample E at 8060 gauss.

Temp. (°C)	ρ (g/cc)	T_1 (sec)
25±2	1.44	160±30
30±2	1.44	150±30
-39±2	2.50	112±10
-47±2	2.58	115±10
-61±2	2.70	90±10
-81±2	2.87	77±4
-101±2	2.98	57±2

given in amagat units. The slope is 2.1 ± 0.4 indicating that T_1 varies as approximately $1/\rho^2$ in this region.

The relaxation time T_1 of Sample E at 25°C was investigated as a function of field. The observed times at 14 500 gauss, 8060 gauss, and in the earth's field were 200 ± 30 sec, 190 ± 40 sec, and 250 ± 60 sec, respectively. Thus no field dependence was observed outside the experimental error.

The Xe¹²⁹ relaxation times for Sample E in the gas phase above 15°C and the liquid phase below this temperature are given in Table II and the product $T_1\rho$ for the liquid phase is plotted in Fig. 4. Again estimated maximum errors are indicated and the magnetic field is 8060 gauss. These relaxation times correspond to points on the coexistence curve, Fig. 2.

At -75°C an exploratory measurement of the diffusion coefficient was made. It was found to be $(0.4 \pm 0.2) \times 10^{-5}$ cm²/sec.

Finally, the most unexpected result of this work was the discovery of large resonant external field shifts in the xenon samples having different densities but a constant resonant frequency.

These shifts are recorded in Table I and plotted in Fig. 5. The field shift is given in terms of $\Delta H = H - H_r$,

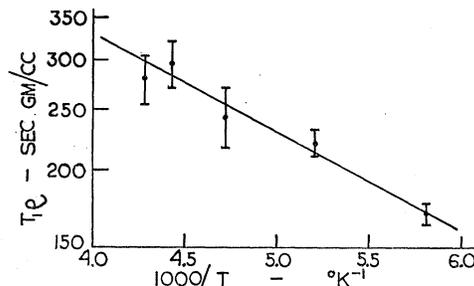


FIG. 4. Semilog plot for the product of the Xe¹²⁹ relaxation time T_1 and the density ρ as a function of reciprocal absolute temperature. The data are for sample E (246 amagats). The magnetic field is 8060 gauss.

where H is the resonant external field for the sample being observed and H_r is the resonant external field for an arbitrary reference sample. This sample, which intentionally contained oxygen, served as a particularly useful reference because of its relatively fast relaxation time. The absolute scale indicating the shift with respect to the reference sample is not important. What is important is the relative shift between two samples with differing densities.

One exploratory observation of the shift was made in the very dense liquid of sample E at -75°C. This shift was -0.53 ± 0.03 gauss and is indicated in Fig. 5. As the density changes from 0.4 g/cc to 2.8 g/cm³, the resonant external field decreases by approximately 1.4 gauss.

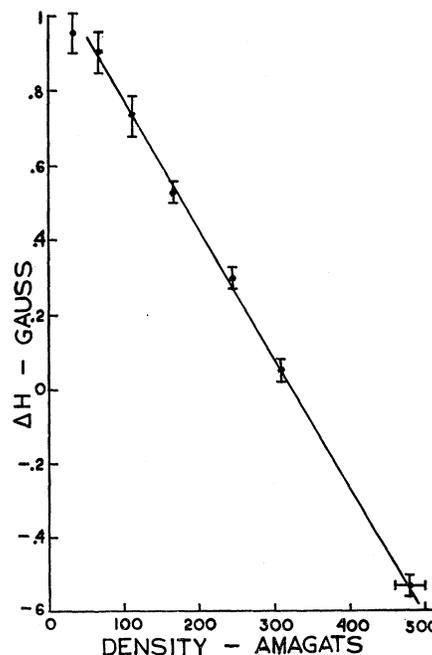


FIG. 5. Resonant external field shift $\Delta H = H - H_r$ at fixed frequency as a function of density. The shifts are given relative to the resonance field H_r of an oxygen-doped reference sample. The total field is approximately 8060 gauss, and all temperatures are 25°C except for the liquid 480-amagat point where the temperature is -75°C.

In order to explore the effect of other molecules on the observed Xe^{129} shift, several samples of approximately 250 amagats were prepared with up to 36 amagats of oxygen. In preliminary measurements the oxygen was found to cause a shift in the Xe^{129} resonance of approximately 11 milligauss/amagat as compared with the 3.45 milligauss/amagat shift due to the xenon.

V. DISCUSSION

Staub and his co-workers^{5,6} measured the room-temperature relaxation time T_1 of Xe^{129} in xenon gas at 50 atm and 74 atm. He obtained 420 sec and 20 sec, respectively. In Table I we report 2600 sec at 48 atm and 160 sec at 73 atm. The origin of these discrepancies is uncertain. If relaxation by impurities is the dominant mechanism, our longer relaxation times are indicative of purer samples.

We have compared our experimental values with theoretical values for two limiting regions, the rare gas and the dense liquid, where relatively good predictions can be made.

For the dense liquid we have used the Bloembergen-Pound-Purcell² theory to calculate T_1 , assuming only a coupling of spin orientations to the nuclear translational motion via direct dipolar magnetic interactions between the nuclear moments. In these "order-of-magnitude" calculations the factors accounting for the presence of two different isotopes¹² or for the finer details of the diffusion process^{3,4} might be omitted. In our calculations we have included the first of these, although not the second which involves less certain assumptions. The high-temperature limit of the expression for T_1 is appropriate for xenon in our 8060 gauss field at -75°C . We obtain a predicted Xe^{129} relaxation time of 8200 sec. Unfortunately, the estimated error in this calculation is approximately 50% pending a more accurate measurement of the diffusion coefficient. But it is sufficiently accurate to indicate that the experimental value of T_1 , 80 ± 10 sec, is approximately two orders of magnitude smaller than the predicted value. The direct nuclear dipole interaction is clearly not the cause of the relaxation.

A somewhat similar circumstance exists in the case of the gas. Xenon behaves approximately as an ideal gas up to 30 atm. We would like to compare experimental and theoretical relaxation times within this region. A 30-atm sample was prepared, but the nuclear signal was too weak and the relaxation time too inconveniently long to permit a good measurement of T_1 . The value of T_1 at 48 atm, for example, is already 2600 ± 600 sec. However, if we extrapolate the data shown in Fig. 3 to 30 atm, we obtain a value 1.1×10^4 sec. Using the Bloembergen¹ rare gas theory based on a direct nuclear magnetic dipole interaction during collisions, we obtain a predicted value for T_1 at 30 atm of 1.1×10^7 sec. Here the theoretical prediction differs from the

experimental value by three orders of magnitude. Again, even allowing for the admittedly crude model of the collision, it is clear that a mechanism other than the direct dipole interaction must be responsible for the relaxation.

In addition to the above two comparisons of absolute values, our data permits us to compare the experimental and theoretical T_1 temperature dependence in the liquid and density dependence in the gas. The Bloembergen-Pound-Purcell² liquid theory indicated that $1/T_1$ is proportional to the density ρ divided by the diffusion coefficient D . One expects the temperature variation of D to be $\exp(-E/RT)$, where T is the absolute temperature and R the gas constant. As shown in Fig. 4, a similar temperature variation has been observed for $T_1\rho$ in our liquid xenon samples. The temperature dependence of the Xe^{129} relaxation time appears to be that of the theory even though the relaxation mechanism is not the direct dipole interaction. Our data for $T_1\rho$ are consistent with an activation energy E of 0.7 ± 0.1 kcal/mole. The diffusion activation energy for xenon has not been measured previously, but a value 0.52 kcal/mole has been reported for liquid argon.¹³

The Bloembergen¹ rare gas theory indicates that $1/T_1$ is proportional to the density ρ . From Fig. 3 it is seen that for xenon between 50 and 100 atm, $1/T_1$ varies approximately as ρ^2 . One should not, however, expect the rare gas theory to be applicable for these pressures above 30 atm. The experimental data in this region indicate a density dependence more consistent with the liquid theory where $1/T_1$ varies as ρ/D .

Having established that the relaxation mechanism is not a direct nuclear magnetic dipole interaction, one must ask if the unknown mechanism might be related to an impurity, particularly oxygen, either within the sample fluid or on the container walls. Using the estimated values given in Sec. II for our oxygen impurity within the fluid, and extrapolating from observed values of T_1 for xenon samples to which controlled amounts of oxygen have been added, we conclude that the relaxation time due to oxygen in our samples is 10^4 sec. Thus we do not believe that oxygen is responsible for the relaxation times reported in Table I. It is also significant to note that our method of sample preparation is such that each sample should contain the same amount of oxygen independent of the amount of xenon. As a result the observed dependence of the gas relaxation time on xenon density is indicative of some relaxation mechanism other than oxygen in the sample.

We also conclude that our relaxation is not caused by magnetic impurities on the walls. This is particularly evident in our liquid samples where, for example, at -75°C it takes an atom approximately 5000 sec to diffuse from the center of the sample to the walls. This is nearly 100 times longer than the relaxation time. In a

¹² I. Solomon, Phys. Rev. **92**, 962 (1955).

¹³ J. Frenkel, *Kinetic Theory of Liquids* (Dover Publications, New York, 1955).

gas at 30 atm, however, such an atom would only take approximately 10 sec to get to the walls. This is 100 times shorter than the relaxation time, and surface relaxation is a real possibility. Relaxation at the surface can be a complicated mechanism which depends not only on the transit time to the surface but also the resident time on the surface and the nature of the surface impurities. We believe, however, that in our gas samples surface relaxation is still not the dominant mechanism. It seems unlikely that a surface relaxation rate would vary with the square of the density.

The possibility of an indirect electron-coupled nuclear magnetic dipole interaction during atomic collisions has been suggested by Bloembergen.¹⁴ This would be a relaxation mechanism only for collisions between atoms with unlike nuclear spins, and could conceivably be an important mechanism in collisions between heavy atoms such as Xe¹²⁹ and Xe¹³¹. Thus far we have not performed experiments to check this suggestion.

Another factor which would influence the Xe¹²⁹ relaxation appreciably could be the existence of stable diatomic molecules in xenon. No experimental evidence for such molecules is known. Their existence, however, has been predicted by Bernardes and Primakoff.¹⁵ It seems unlikely that the existence of such molecules in appreciable numbers is consistent with the T_1 density dependence observed in our gas samples. In a diatomic gas T_1 is normally directly proportional to the density.¹ It is necessary to take account of the monatomic-diatom mixture,¹⁶ but this does not provide the strong inverse dependence which we have observed in the gas. Our observed density dependence is more nearly consistent with a monatomic gas where T_1 is normally expected to be inversely proportional to the density.

¹⁴ N. Bloembergen (private communication).

¹⁵ N. Bernardes and H. Primakoff, *J. Chem. Phys.* **30**, 691 (1959).

¹⁶ H. C. Torrey (to be published).

It is significant to note that in our 8060-gauss external field, an isolated xenon atom has a strong diamagnetic field of 45 gauss. Our shift may be related to fluctuations of this 45-gauss field during atomic collisions. The sign of our shift is such that a preference for smaller values of the diamagnetic field exists as the density is increased. The shift would be expected to be field dependent, but we have not as yet checked this experimentally.

The relaxation mechanism may also be related to the collisions. A fluctuating magnetic field exists at the nucleus as a result of the motion of the nonsymmetrical electronic charge distributions which exist during collisions. Because of the rapid oscillations of the dipole distributions between two colliding molecules, the intermolecular magnetic field at a given nucleus averages to zero. But the intramolecular field arising from the motion of the nonsymmetrical charge distribution within the xenon atom does not average to zero during a given collision. This may provide a dominant relaxation mechanism for Xe¹²⁹. Using this mechanism Torrey¹⁶ has calculated absolute values of T_1 , the field dependence of T_1 , and a Korringa-type relationship between T_1 and ΔH . He has obtained good agreement with our experimental observations.

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