

Hyperfine Relaxation of Optically Pumped Rb^{87} Atoms in Buffer Gases

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The hyperfine relaxation of a population difference, produced by optical pumping, between the $F=1$ $m_F=0$ and $F=2$ $m_F=0$ energy levels of the ground state of Rb^{87} , has been studied in cells containing neon, argon, helium, or krypton as a buffer gas. The relaxation times T_1 and T_2 have been measured as a function of the rubidium vapor pressure in the cell. The relaxation rate due to spin exchange in Rb^{87} - Rb^{87} collisions has been found equal to 65 sec^{-1} at 44°C , and the diffusion coefficients of Rb^{87} atoms in He, Ne, and Ar evaluated as 0.6, 0.31, and $0.22 \text{ cm}^2 \text{ sec}^{-1}$, respectively. The cross section for Rb^{87} -Ne and Rb^{87} -Ar collisions are found to be $1.6 \times 10^{-22} \text{ cm}^2$ and $9 \times 10^{-22} \text{ cm}^2$. These values are in general agreement with values previously obtained by other methods. The extrapolated values of $1/T_2$ for zero vapor pressure of rubidium are 185, 237, and 318 sec^{-1} for Ne, He, or Ar, respectively. Relaxations by the pumping light or by the magnetic field inhomogeneities are also discussed.

I. INTRODUCTION

SPIN relaxation of alkali metal vapors in the presence of inert buffer gases has been previously studied,¹⁻⁴ and more specially in the case of rubidium atoms. However, what has been measured principally was the relaxation time of the mean electronic polarization $\langle S_z \rangle$ of the rubidium atoms, averaged over all the sub-levels of the ground state, and in this case, the relaxation due to spin-exchange collisions between alkali atoms was not considered important. There has been also some ambiguity in the exact determination of the relaxation times T_1 and T_2 .

It has been suggested⁵ that a more suitable experiment is one employing hyperfine population difference, the observable $\langle \mathbf{S} \cdot \mathbf{I} \rangle$, and unpolarized light. Such a method has been utilized recently⁶ for rubidium atoms in carefully evacuated cells with wall coatings, and it has been found in that case that the spin-exchange collisions were the dominant factor in the relaxation process for T_1 . In this paper the method has also been applied to the study of the hyperfine relaxation of rubidium atoms in a cell containing a buffer gas. The experimental arrangement is different from the previous works and permits the measurements of T_1 and T_2 . The value of the spin-exchange relaxation obtained by these measurements agrees closely with the value obtained by Bouchiat and Brossel. The effects produced on T_1 and T_2 by the collisions of the rubidium atoms with the walls or the buffer-gas atoms will be discussed in the case of neon, argon, helium and krypton.

II. RELAXATION PROCESSES

In many double resonance experiments using optical pumping of the alkali atoms in the ground state, non-magnetic buffer gases are used both to reduce the Doppler width of the magnetic resonance⁷ and to increase the efficiency of optical pumping.⁸ Since buffer gases are very effective in reducing the Doppler broadening, other relaxation processes become predominant. Such processes include principally spin-exchange collisions between alkali atoms,⁹ wall collisions, pressure broadening due to collisions between alkali atoms and buffer gas atoms, broadening due to magnetic field inhomogeneities, and broadening due to relaxation by the pumping light.

In the so-called "static" method of measurement, where the resonance curve is plotted as a function of

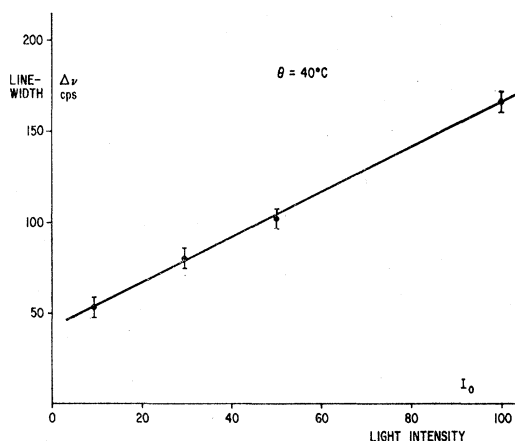


FIG. 1. Variation of linewidth of 0-0 transition in Rb^{87} versus the intensity of pumping light under continuous illumination.

¹ H. G. Dehmelt, *Phys. Rev.* **105**, 1487 (1957).

² W. Franzen, *Phys. Rev.* **115**, 850 (1959).

³ R. A. Bernheim, *J. Chem. Phys.* **36**, 135 (1962).

⁴ L. W. Anderson, F. M. Pipkin, and J. C. Baird, Jr., *Phys. Rev.* **116**, 87 (1959). L. C. Balling, R. J. Hanson, and F. M. Pipkin, *Phys. Rev.* **133**, A607 (1964).

⁵ A. L. Bloom, *Phys. Rev.* **118**, 664 (1960).

⁶ M. A. Bouchiat and J. Brossel, *Compt. Rend.* **257**, 2825 (1963).

⁷ R. H. Dicke, *Phys. Rev.* **89**, 472 (1953).

⁸ A. Kastler, *J. Opt. Soc. Am.* **47**, 460 (1957).

⁹ J. P. Wittke and R. H. Dicke, *Phys. Rev.* **103**, 620 (1956); R. M. Mazo, *J. Chem. Phys.* **34**, 169 (1961).

the frequency, it is not easy to distinguish the contributions of the various relaxation processes by a direct determination of the linewidth of the resonance curve. Extrapolation methods where only one factor is varied at a time, such as the intensity of the pumping light (Fig. 1) or the value of the rf magnetic field, are time consuming and uncertain.

Substituting time-domain measurements instead of frequency-domain measurements, the "dynamic" methods for the determination of the relaxation times are based on the observation of the transients taking place when sudden changes are produced in the pumping-light intensity or in the rf magnetic field.^{2,10} In this latter case, the time constant T of the transient is a function of two relaxation mechanisms, one which changes the magnetization along the axis of quantization, and another which causes a loss of coherence between the oscillating moment and the rf field. By analogy with the case of nuclear magnetic resonance, "longitudinal" and "transverse" relaxation times T_1 and T_2 are used to distinguish the decay rates due to each of these two processes.

Franzen's method permits the measurement of T_1 in $\langle S_z \rangle$ and eliminates the effect of the relaxation due to the pumping light by allowing an aligned vapor to relax in the dark. A diffusion theory was developed, taking into consideration the depolarization of the rubidium atoms by collisions with the walls and by collisions with the buffer-gas atoms, and neglecting the spin-exchange collisions effect. The experimental data obtained with natural rubidium and neon, argon,² or helium³ agreed well with the theoretical predictions. The relaxation time measured was given by

$$T_1 = [kD_0(p_0/p) + N_0\sigma_1\bar{v}_{rel}(p/p_0)]^{-1}, \quad (1)$$

where D_0 is the coefficient of diffusion for rubidium atoms in the buffer gas at atmospheric pressure p_0 ; $D_0(p_0/p)$ is the value of the diffusion coefficient at the actual pressure p ; σ_1 is the disorientation cross section which characterizes rubidium-buffer gas collisions; N_0 is the density of buffer gas atoms at atmospheric pressure p_0 , while $N_0(p/p_0)$ is the density at pressure p . The factor k contains the dimensions of the actual cell used in the experiments.³

More recently Bouchiat and Brossel have carefully studied the relaxation in the observable $\langle \mathbf{S} \cdot \mathbf{I} \rangle$, for rubidium atoms in evacuated cells with special wall coatings, using hyperfine pumping with nonpolarized D_1D_2 lines. In such a case the rubidium vapor is not polarized $\langle S_z \rangle = 0$, but a large population difference $\langle \mathbf{S} \cdot \mathbf{I} \rangle$ takes place between the $F=1$ and $F=2$, levels of the ground state. Using Franzen's method of relaxation in the dark, the value of T_1 was measured as a function of the rubidium-vapor pressure. Here, the relaxation of $\langle \mathbf{S} \cdot \mathbf{I} \rangle$ is produced only by wall collisions and by spin-

exchange collisions between rubidium atoms. A theory¹¹ shows that wall relaxation should produce an exponential decay rate with a time constant T_W . Similarly, it has been established theoretically,¹² that when $\langle S_z \rangle = 0$, the spin-exchange collisions produce also an exponential decay rate of $\langle \mathbf{S} \cdot \mathbf{I} \rangle$ with a time constant T_{EX} such that

$$1/T_1 = 1/T_W + 1/T_{EX}. \quad (2)$$

Experiments have confirmed these theoretical predictions and a value of 6 to 8×10^{-14} cm² has been quoted⁶ for the spin-exchange cross section of $\text{Rb}^{87}-\text{Rb}^{87}$ or $\text{Rb}^{85}-\text{Rb}^{85}$ or $\text{Rb}^{87}-\text{Rb}^{85}$ collisions, with most of the uncertainty in the estimate of the rubidium vapor pressure from temperature measurements. This cross section is somewhat higher than the value of $(1.7 \pm 0.21) \times 10^{-14}$ cm² obtained by Jarrett¹³ for $\text{Rb}^{87}-\text{Rb}^{85}$ spin-exchange collisions, with a different experimental method.

In the present paper, a method is described for the measurement of T_1 and T_2 deduced also from the exponential decay of $\langle \mathbf{S} \cdot \mathbf{I} \rangle$ for rubidium atoms optically pumped in a gas cell. The results of these experiments will be compared with the values quoted in the works previously described.

III. EXPERIMENTAL PROCEDURE

To reduce the effects of magnetic-field inhomogeneities, the exponential decay of a population difference, initially created by hyperfine pumping, between the $F=1m_F=0$ and $F=2m_F=0$ levels in the ground state of rubidium atoms was chosen for this study. Monitoring of the pumping-light intensity transmitted through the cell, however, is not always easy to interpret in terms of population changes between these two levels, especially when the vapor pressure of the rubidium atoms is varied. For this reason it has been preferred to monitor the microwave-induced emission produced after a pulse of microwave energy, at the resonance frequency, is applied to the cell in a microwave cavity.

The method is similar to the pulsed decay methods which have been applied to nuclear resonance relaxation studies in the past.¹⁴ The rubidium atoms are placed in a radiative state by a short pulse of microwave at the resonance frequency; at the end of this pulse, a signal, at the resonance frequency, is generated in the cavity by the rubidium atoms, and the amplitude of this signal decreases with a decay constant T , which is measured.

To obtain separately T_1 and T_2 the following procedure is observed. First, a large population difference between the two 0-0 hyperfine levels is created by a strong pulse of resonance light. Second, a microwave pulse is then applied at a later time Δt , and immediately

¹¹ M. A. Bouchiat, J. Phys. Radium **24**, 379 and 611 (1963).

¹² F. Grossetete, J. Phys. Radium (to be published).

¹³ S. M. Jarrett, Phys. Rev. **133**, A111 (1964).

¹⁴ E. L. Hahn, Phys. Rev. **76**, 145 (1949); **77**, 297 (1950).

¹⁰ B. Cagnac and J. Brossel, Compt. Rend. **249**, 253 (1959); B. Cagnac Ph.D. thesis, Paris, 1961 (unpublished).

following this pulse, an induced signal appears. This signal decays exponentially with a time constant T , but the initial amplitude of the signal is an indication of the population difference still existing between the two 0-0 levels, at a time Δt after the end of the light pulse. In this method the relaxation also takes place in the dark, and T_1 can be obtained by comparison of initial decay amplitudes for various known times Δt (Fig. 2). By analogy with somewhat similar experiments,¹⁰ T_2 can be obtained from the relation

$$2/T = 1/T_1 + 1/T_2. \quad (3)$$

This equation, in the context of the experimental method, defines T_2 as the transverse relaxation rather than the inverse linewidth, although in this work we do not measure the linewidth directly.

When applying this method some precautions must be taken: First, it is necessary to limit the minimum time Δt between the end of the light pulse and the beginning of the microwave pulse, so that the population change is fully established following the end of the light pulse (this time is usually of the order of 1 msec); second, the duration of the probing microwave pulse should be very short compared with T_1 , and the amplitude of the microwave field small enough not to saturate the transition.

These conditions somewhat reduce the S/N of the detection of the pulse induced microwave signal. For this reason the method is not easily applicable to situations where T_1 is smaller than about 2 msec, as in krypton or xenon. However, the method has been applied with success to the measurements of the relaxation times of Rb⁸⁷ in neon, argon, and helium.

The experimental arrangement is shown in Fig. 3.

A glass cell containing Rb⁸⁷ and a nonmagnetic buffer gas is enclosed in a microwave resonant cavity operating in the TE_{011} mode. The cell is optically pumped with unpolarized light coming from a Rb⁸⁷ lamp and filtered through a Rb⁸⁵ cell also containing a buffer gas to broaden the absorption line and to increase the efficiency of the hyperfine filtering. In this case, the hyperfine pumping favored the population of the upper level

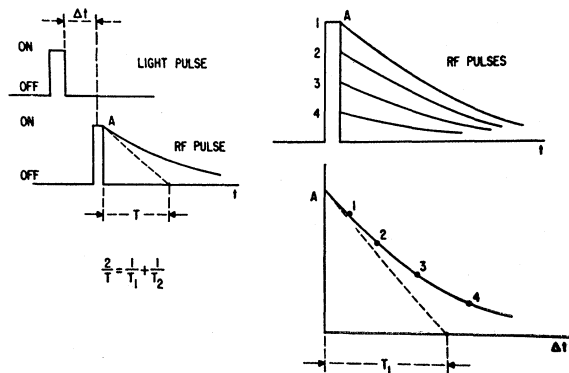


FIG. 2. Principle of the method of measurement of T_1 and T_2 .

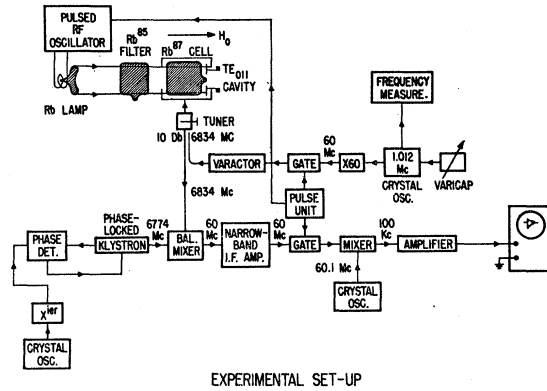


FIG. 3. Experimental setup for measurements of hyperfine relaxation.

$F = 2m_F = 0$. On the other hand, the population of the lower level $F = 1m_F = 0$ can be favored by pumping the Rb⁸⁷ sample cell directly with the light from a lamp containing pure Rb⁸⁵, without any filtering.¹⁵ In both cases, a pulse-induced emission signal can be obtained after the removal of the exciting microwave pulse.

The sample cell, in the cavity, and the filter cell are thermostatically controlled. The rubidium lamp is excited by a pulsed rf oscillator and a pulse-control unit permits the light pulse to be adjusted in time with respect to a reference pulse.

The microwave frequency is obtained by multiplication from a stable crystal oscillator up to 60+ Mc, and harmonic generation by a varactor diode up to 6834 Mc. A gated amplifier, in series with the multiplier chain, permits the microwave energy to be delivered in pulses, at various time intervals with respect to the light pulse. In a weak magnetic field, by applying a pulse of microwave at the resonance frequency, the atoms will continue to radiate for some time after the removal of the microwave pulse. The detection of this pulse-induced emission signal is obtained through a microwave superheterodyne receiver coupled to the reflection cavity. A double detection system reduces the over-all bandwidth of the receiver and increases the S/N of the detection. To avoid saturation of the receiver, a gated stage in the i.f. amplifier turns off the receiver during the time that the microwave pulse is sent into the cavity. The bandwidth of the amplifiers is large enough so that the time constants of the circuits do not interfere with the measurement of the exponential decay of the microwave-induced emission from the rubidium atoms. For accurate determinations of T_1 and T_2 , pictures were taken of the oscilloscope display of the detection of the pulse-induced emission signal. A

¹⁵ Hyperfine pumping was suggested independently by Prof. A. Kastler, Ecole Normale Supérieure Paris; Dr. P. L. Bender, National Bureau of Standards, Washington; and Professor T. R. Carver and C. O. Alley, Jr., Princeton University in 1958. See also P. Bender, in *Quantum Electronics*, edited by C. H. Townes (Columbia University Press, New York, 1960), p. 115.

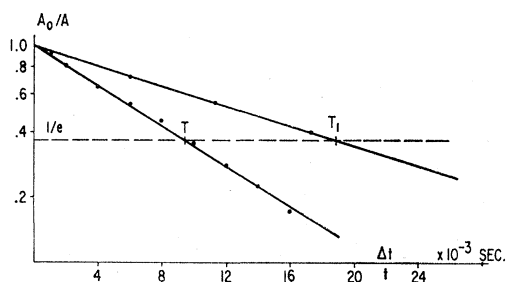


FIG. 4. Typical plot of $\log A_0/A$ versus Δt . From the above plot $T_1 = 19 \times 10^{-3}$ sec and $T = 9.3 \times 10^{-3}$ sec.

semilogarithm plot of the ratio of the initial amplitudes of the exponential decay A_0/A against time is shown in Fig. 4, from which the relaxation time is evaluated as the slope of the best straight line through the experimental points (A_0 corresponds to the initial amplitude of the decay signal for Δt close to zero).

IV. EXPERIMENTAL RESULTS

The measurements were made with sealed-off cells made of 707 glass filled with pure Rb⁸⁷ (99.7% pure) and spectroscopically pure buffer gases at various pressures: neon at 5, 10, and 50 Torr, argon at 5 and 30 Torr, helium at 14 Torr, and krypton at 10 Torr. To see the effect of the walls, three cells filled with 10 Torr of neon were made—one of 707 glass, another of 707 glass coated with GE SC-77 Dri-film, a mixture of dimethyl-dichlorosilicane and methylchlorosilicane,¹⁶ and another of quartz.

Preliminary to the measurements, the effect of the microwave saturation was determined in order to stay within the limits previously mentioned to minimize the perturbations by the probing microwave field on the measurements of T_1 . For example, the relaxation times were relatively constant until a power level capable of saturating the resonance was approached. Increasing the microwave power above this point causes the measured relaxation time T_1 to drop considerably (Fig. 5). Also within these limits of power level, the duration of the microwave pulse was not critical and did not affect the measurements of T_1 . When these precautions were taken, a single exponential time dependence was observed for the induced signal and also for the curve of the initial amplitude of the decay signal versus the elapsed time Δt between the light pulse and the microwave pulse, thus justifying an expression for T_1 analogous to Eq. (2).

With a cell filled with 10 Torr of neon, no difference was observed in the measurements of T_1 and T_2 when the hyperfine pumping favored the population of either the upper energy level or the lower energy level. For purposes of comparison, all the measurements were

then made subsequently with hyperfine pumping preferentially populating the upper energy level.

The microwave cavity was well thermostated and when varying the temperature sufficient time was allowed for the sample cell to reach thermal equilibrium. However, there is some uncertainty in determining the vapor pressure of the rubidium atoms from a temperature measurement of the cell. The pressure of the saturated vapor¹⁷ at the temperature T is given by

$$\log_{10} p = -4560/T + 12.00 - 1.45 \log_{10} T. \quad (4)$$

This expression at 90°C gives a value of p about five times higher than the value measured by Jarrett¹³ with an interferometric method of density measurement, recently developed by Jarrett and Franken,¹⁸ and felt to be capable of yielding results of about 10% accuracy. However, their method has been applied only to one value of the temperature. We do not here attempt to provide an absolute value for the spin-exchange cross section, but have used the expression (4) as a measure of the temperature dependence of the vapor pressure. Experimentally the spin-exchange relaxation rate is found to be linear with p as deduced from (4), and its use is convenient for extrapolation to zero vapor pressure, even though it is presumably of little value for the determination of an absolute spin-exchange cross section.

1. Measurements of T_1

The results are resuméd in Fig. 6 where $1/T_1$ has been plotted as a function of the vapor pressure of the rubidium atoms. The relaxation rate $1/T_1$ appears to be a linear function of the number N of rubidium atoms and, since $1/T_{EX} \propto N$, an expression similar to Eq. (2)

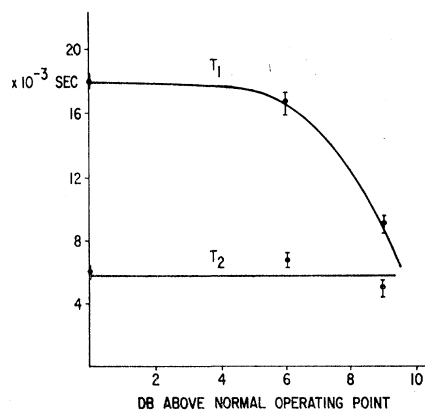


FIG. 5. Decrease in relaxation time T_1 as a function of the exciting microwave power level.

¹⁷ *Metals Reference Handbook*, edited by Colin J. Smithells (Butterworths Scientific Publication Ltd., London, 1962), Vol. II, p. 655.

¹⁸ S. M. Jarrett and P. A. Franken (to be published).

¹⁶ C. O. Alley, Jr., Final Report, U. S. Army Signal Corps Engineering Laboratory, 30 September 1960 (unpublished).

TABLE I. Diffusion coefficient D_0 and cross section σ_1 for rubidium atoms in various buffer gases.

Buffer gas	D_0 (cm ² /sec)	σ_1 (cm ²)
He	0.6	
Ne	0.31	1.6×10^{-22}
Ar	0.22	9×10^{-22}

can be justified for the case of rubidium cells filled with a buffer gas:

$$1/T_1 = 1/T_{EX} + 1/T_G, \quad (5)$$

where $1/T_{EX}$ represents the relaxation due to spin-exchange collisions and $1/T_G$ the relaxation due to collisions between the rubidium atoms and the buffer-gas atoms or the walls.

The extrapolated value of $1/T_1$ for zero vapor pressure corresponds to $1/T_G$, independent of the vapor pressure. For the various buffer gases the lines are all parallel and there is very little difference between them because, as the vapor pressure increases, $1/T_{EX}$ becomes very rapidly the dominant factor in Eq. (5). It is interesting also to note the parallelism of these lines with the line extrapolated from data obtained by Bouchiat and Brosset for evacuated and coated cells, for a much shorter range of vapor pressure. Whereas the spin-exchange collisions were not considered significant in T_1 , as deduced from the decay of the polarization $\langle S_z \rangle$, they are preponderant in T_1 as deduced from $\langle S \cdot I \rangle$.

The extrapolated values of $1/T_G$ for zero vapor pressure are 23, 37, and 42 sec⁻¹ for neon, helium, and argon, respectively, and in the case of neon and argon, where such checks were made, seem to be independent of the pressure of the buffer gas within the range of pressure

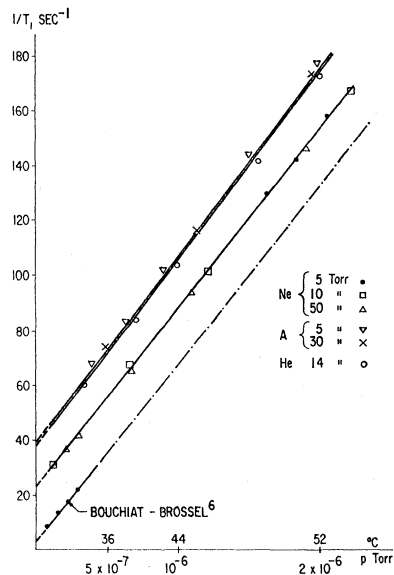


FIG. 6. Relaxation rate $1/T_1$ versus rubidium-vapor pressure for various buffer gases.

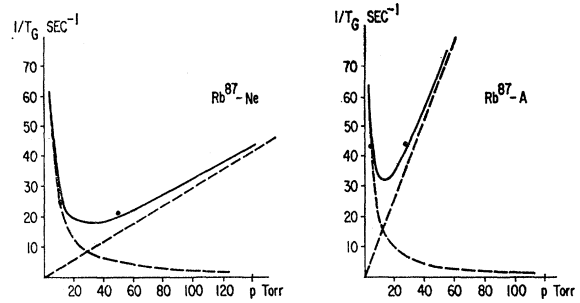


FIG. 7. Relaxation rate $1/T_G$ versus gas pressure for neon and argon.

from 5 to 50 Torr. These results can be explained by the simple diffusion theory proposed earlier by Franzen,² where it is assumed that the wall collisions are non-adiabatic and that relaxation occurs during each single collision of the rubidium atoms with the walls of the cell.

Referring to Eq. (1) the experimental data are best fitted by taking $k = 1.1$ cm⁻² for a spherical cell of about 2 in. diam. and for the values of D_0 and σ_1 , shown in Table I.

The values of D_0 for neon, argon and helium shown in Table I agree well with the values quoted by Franzen and Bernheim^{2,3}; the values of σ_1 , however, are larger by a factor of 3 in the case of neon or argon. In the case of helium the small value of σ_1 makes it difficult to see the contribution of this factor, in the range of pressures of the experiments, whereas in the case of krypton σ_1 is so large and the relaxation times so short (smaller than 2 to 3 msec) that the method could not be applied with any accuracy.

In the case of neon and argon, Fig. 7 shows the respective contributions of the two factors in $1/T_G$ in Eq. (1), as the pressure of the buffer gas is varied, resulting in an almost constant value of $1/T_G$ in the range of pressures from 5 to 50 Torr, as found also experimentally.

The experimental results were the same for cells made of different wall materials. A contamination of the walls by deposited rubidium atoms may be the explanation for such uniformity of results.⁶

2. Measurements of T_2

The relaxation rate $1/T_2$ is due to adiabatic collisions during which no transition of the atoms between its states are induced but in which a small change in the spacing of the energy levels usually occurs, leading to a loss of coherence of the induced microwave signal due to the randomness of the perturbations.¹⁹

From the measurements of T and T_1 , T_2 can be deduced from Eq. (3). First, it has been checked that T_2 , unlike T_1 , was independent of the amplitude of the

¹⁹ D. Kleppner, M. Goldenberg, and N. F. Ramsey, Phys. Rev. 126, 603 (1962).

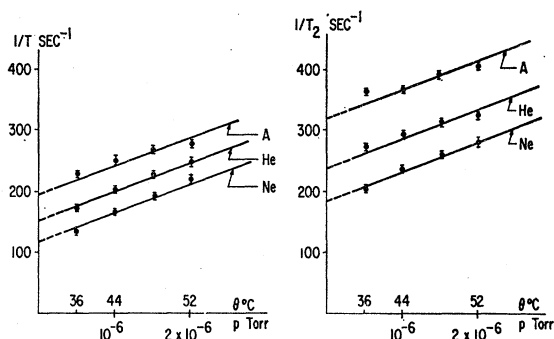


FIG. 8. Relaxation rate $1/T_2$ versus rubidium vapor pressure for various buffer gases.

exciting microwave pulse (Fig. 5). Second, the dependence of T_2 on the rubidium vapor pressure was measured for cells filled with argon, neon or helium. The experimental results are shown in Fig. 8. Here again, a set of parallel lines have been obtained when plotting $1/T_2$ versus the rubidium vapor pressure, and the slope of these lines is the same as the slope of the lines representing $1/T_1$ versus rubidium vapor pressure. This indicates that spin-exchange collisions are also a contributing factor to the value of T_2 and justify an expression such as

$$1/T_2 = 1/T_{\text{EX}} + 1/T_{G'}, \quad (6)$$

where $1/T_{G'}$ represents the relaxation rate due to adiabatic rubidium-wall collisions and rubidium-buffer gas collisions. The rate $1/T_{G'}$ is constant with temperature and can be obtained by extrapolating the values of $1/T_2$ to zero vapor pressure.

It is shown in Ref. 19, and from a simple random walk argument, that the radiative atom loses coherence after a number of collisions:

$$n \approx 2/\phi^2, \quad (7)$$

where ϕ is the phase shift per collision, and if the collision rate is $1/t_c$ it follows that

$$1/T_{G'} = \frac{1}{2}(\phi^2/t_c). \quad (8)$$

The phase shift is related to the frequency shift Δf produced by the walls or the buffer gas by

$$\Delta f = \phi/2\pi t_c. \quad (9)$$

The frequency shifts produced by buffer gas in the 0-0 transition of Rb^{87} have been measured.²⁰ However, accurate data on the frequency shifts produced by glass walls are missing, and for this reason it has not been possible to check quantitatively the values of $1/T_{G'}$ in a manner similar to $1/T_G$.

The values of $1/T_{G'}$ are 185, 237, and 318 sec^{-1} for neon, helium, and argon, respectively, and $1/T_{G'}$ is again constant for neon and argon in the range of pres-

sure from 5 to 50 Torr. By subtracting the effect of spin-exchange collisions from the values of T_1 and T_2 , the resulting difference is due only to the difference between T_G and $T_{G'}$, with $1/T_{G'}$ about 8 times larger than $1/T_G$. These measurements permit a better understanding of the comparative values of T_1 and T_2 . The values of T_1 and T_2 approach each other only when the temperature of the cell is so high that the spin-exchange collisions are the dominant factor.

3. Relaxation by the Pumping Light

In the experiments previously described, the pumping light was applied as a pulse and the relaxation observed in the dark, so that the relaxation by the light was eliminated. In another experiment, intended to study the effect of the relaxation produced by the light, the cell was continuously illuminated, under conditions of hyperfine pumping, but the microwave was still applied as a short pulse. The exponential decay T' of the microwave-stimulated emission signal after the removal of the microwave pulse was again studied as a function of the temperature of the cell and for various intensities of the pumping light. This experiment was carried on with a cell containing 10 Torr of neon.

Under these conditions, the microwave excitation placed the rubidium atoms in superposition states from which they could be driven off by two random processes: One of them is the thermal relaxation as previously defined, and the other is the excitation by light to excited states from which they fall back to the pure states $F=1$ or $F=2$. These two processes are random and lead to time-independent relaxation rates T and T_L , so that the total relaxation rate is the sum of the rates for each process:

$$1/T' = 1/T + 1/T_L. \quad (10)$$

Figure 9 shows the values of $1/T'$ as a function of the rubidium-vapor pressure and for different light intensities I_0 . Here again the lines are parallel and their slope is the same as the slope of $1/T_1$ versus vapor pressure as shown in Fig. 6. The relaxation due to the pumping light is directly proportional to the light intensity and, by extrapolating to zero light intensity, one can obtain a curve of $(1/T')_{I=0}$ versus vapor pressure similar to the curve of $1/T$ obtained previously by the pulsed-light method and shown in Fig. 8. By extrapolation to zero vapor pressure a value of 150 sec^{-1} is obtained for $1/T_L$ at full light intensity.

4. Effect of Magnetic Field Inhomogeneities

A strong field inhomogeneity in the cell was produced by a small permanent magnet located near a cell containing 10 Torr of neon. It was observed in that case that the relaxation time T_1 was not noticeably changed, but that the relaxation time T and consequently T_2 was changed considerably. The effect of field inhomogeneity

²⁰ P. L. Bender, E. C. Beaty, and A. R. Chi, Phys. Rev. Letters **1**, 311 (1958).

generality on T_1 is primarily in transferring atoms from the $m_F=0$ state to the other Zeeman levels with the same F value, and in accordance with theory is considerably longer at the value of H_0 used. The rate $1/T_2'$ changed from 225 to 540 sec^{-1} at a temperature of operation of 40°C.

Writing

$$1/T_2' = 1/T_2 + 1/T_M, \quad (11)$$

where $1/T_2$ corresponds to a uniform magnetic field and $1/T_M$ corresponds to the field inhomogeneities, and neglecting the effect of the inhomogeneities on the Zeeman states but estimating the phase decorrelation due to random motion through an inhomogeneous field, according to the theory developed in Ref. 19,

$$1/T_M = t_0(16\alpha^2 H_0^2 \Delta H^2), \quad (12)$$

where H_0 is the static field, α the field-dependent coefficient of the $F=1m_F=0$ to $F=2m_F=0$ transition frequency ($\alpha=574$ cps for Rb⁸⁷), ΔH the total field inhomogeneity across the cell, and t_0 the time it takes an atom to cross the length of the bulb in the direction of H_0 .

With $t_0=0.03$ sec, $\alpha=574$ cps, $H_0=0.4$ Oe, and $\Delta H=0.1$ Oe, we find $1/T_M=256$ sec^{-1} in agreement with experimental values.

5. Application to Rubidium Maser

The threshold condition²¹ for Maser oscillation can be expressed by

$$n_2 - n_1 > h(4\pi^2 T Q_0 \mu^2)^{-1}, \quad (13)$$

where $n_2 - n_1$ is the number of excess atoms in the population of the upper energy level, Q_0 the unloaded Q of the cavity, and T the relaxation time of the atoms.

For ease of operation of the maser, T should be as large as possible. From the previous measurements it can be seen that the relaxations produced by the buffer gas and the walls are not negligible in limiting the value of T and that a greater relaxation time could be obtained when using specially coated cells and no buffer gas.⁶ However, even in this case, at the normal tempera-

²¹ A. L. Schawlow and C. H. Townes, Phys. Rev. **112**, 1940 (1958).

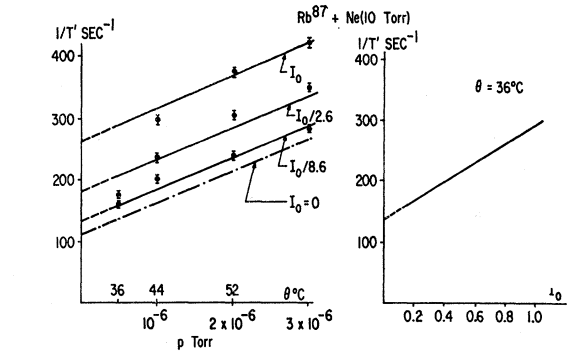


Fig. 9. Effect of intensity of pumping light on relaxation rate $1/T'$ versus rubidium-vapor pressure (continuous illumination).

ture of operation necessary to obtain a significant value of $n_2 - n_1$, say 44°C for example, the spin-exchange collisions will limit the maximum value of T_1 to about 14 msec, and the resulting relaxation time T to an even lower value. This is to be compared with a relaxation rate of the order of 10^{-4} sec due to spin-exchange in hydrogen-hydrogen collisions. An over-all relaxation time of about 1 sec has been obtained with the hydrogen Maser using spin-state selector and Teflon coated bulb in the microwave cavity.¹⁹

The strong effect of spin-exchange collisions in rubidium atoms is one of the main difficulties in the realization of a self-oscillating rubidium maser. However, a regenerative rubidium Maser oscillator, locked to the 0-0 microwave hyperfine transition, has been previously described²² and more recently a successful realization of a self-oscillating rubidium maser has been announced.²³

V. CONCLUSIONS

We believe that the ability of pulse methods to separate out the effects of different relaxation processes adds considerably to the total picture of relaxation in optical pumping, even though the separate effects have been studied in separate experiments in the past.

²² M. Arditi and T. R. Carver, Bull. Am. Phys. Soc. **9**, 65 (1964).

²³ P. Davidovits, Appl. Phys. Letters **5**, 15 (1964).