PHYSICAL REVIEW A VOLUME 16, NUMBER 2 AUGUST 1977

Spin relaxation of optically pumped cesium in high magnetic fields*

T. R. Marshall,† R. Boggy, and F. A. Franz

Department of Physics, Indiana University, Bloomington, Indiana 47401

(Received 17 February 1977)

Cs vapor in He, Ne, and Ar buffer gases has been optically pumped in magnetic fields as high as 100 kG. The rate of electronic spin relaxation measured in Cs-Ar is approximately the same as that predicted by the low-field nuclear-spin-independent cross section for the relaxation of $\langle S_z \rangle$ via the $\gamma(\vec{S}\cdot\vec{N})$ (spin-orbit) interaction. In the cases of Cs-He and Cs-Ne, considerably higher relaxation rates are measured at high fields than those that are predicted from the low-field cross sections. The differences are shown to be due to relaxation arising from the collision-induced modification of the Cs hyperfine interaction, $\delta a(\vec{S}\cdot\vec{I})$. Upper limits of the order of 10^{-13} sec are set for the average durations of both the $\delta a(\vec{S}\cdot\vec{I})$ and the $\gamma(\vec{S}\cdot\vec{N})$ interactions in sudden binary collisions of Cs atoms with light noble-gas atoms. The average cross sections for the $\delta a(\vec{S}\cdot\vec{I})$ interactions are estimated to be 1.3×10^{-14} cm^2 for Cs-He, and 0.73×10^{-14} cm^2 for Cs-Ne.

I. INTRODUCTION

Optical-pumping experiments on alkali-metal vapors usually have been performed under conditions such that the hyperfine interaction between the electronic and nuclear spins of the alkali-metal atom $a(\vec{S}\cdot\vec{I})$ is large compared to the interaction of $\vec{S}$ with any externally applied magnetic field.[1] The nuclear spin then acts as a "reservoir" of spin polarization, effectively "slowing" certain relaxation rates and altering the ground-state $\langle S_z \rangle_g$ relaxation transient. In magnetic fields of the order of 10 kG and above, however, the electronic spin is decoupled from the nuclear spin, and relaxation rates, uninfluenced by the normal low-magnetic-field-nuclear-spin dynamics, should be measurable.

In the present paper, we report measurements of the relaxation of Cs in He, Ne, and Ar buffer gases, at magnetic fields ranging from 20 to 100 kG. The high-field relaxation rates measured for Cs-Ar are almost exactly those that are predicted from low-magnetic-field measurements of nuclear-spin-independent cross sections for collisional relaxation of $\langle S_z \rangle$ via the $\gamma(\vec{S}\cdot\vec{N})$ (spin-orbit) interaction. The high-field relaxation rates for Cs-He and Cs-Ne, however, turn out to be considerably larger than those that are predicted from the low-field cross sections. The difference is shown to be due to contributions—at high magnetic field only—from the collision-induced modification of the alkali-metal-atom–hyperfine interaction $\delta a(\vec{S}\cdot\vec{I})$ the same interaction responsible for the well-known pressure shift of hyperfine frequencies at low magnetic field. By comparing relaxation rates at 100 kG with those at lower fields, we set limits on the correlation times for both the $\delta a(\vec{S}\cdot\vec{I})$ and $\gamma(\vec{S}\cdot\vec{N})$ modes of relaxation in Cs-atom–noble-gas-atom sudden binary collisions, and gain information on the appropriate model describing Cs relaxation on alkali-metal-contaminated glass surfaces.

II. EXPERIMENTAL APPARATUS

The heart of the system shown schematically in Fig. 1 was an optical pumping cell centered in the room-temperature access bore of a superconducting solenoid. A circularly polarized D_1(8944-Å) white-light pumping beam passed upward through the cell, while a weak D_2(8521-Å) white-light detection beam passed downward. Monitoring of the detection beam permitted observation of transient changes in $\langle S_z \rangle_g$ induced by turning the pumping beam on and off.[2] Suitably averaged transient signals were analyzed by computer, and provided the data which serve as the bases for this paper. Each component of the experimental system is described in more detail below.

A. Magnetic field

The magnetic field was supplied by an RCA superconducting solenoid capable of reaching 100 kG, with a room-temperature access bore of 3.2 cm. The field homogeneity over the 2.5-cm-long optical pumping cell was approximately 10^{-4}.

B. Optical pumping cells

The optical pumping cells were uncoated Pyrex cylinders, 2.6-cm internal diameter, 2.5-cm internal length, with a small tubular "pull-off" blown onto the edge of one face, as shown in Fig. 2. The cells were prepared on a bakable all metal and glass gas handling system into which Cs was dis-

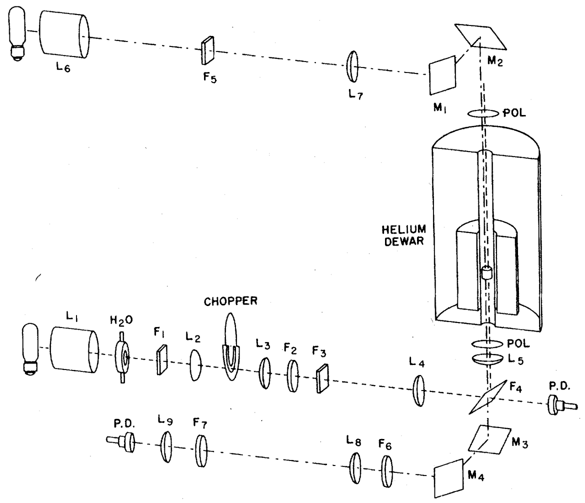

FIG. 1. Schematic of the high-field optical pumping apparatus. The D_1 (chopped) pumping beam originates at the bottom, passes horizontally from left to right, then upward through the optical pumping cell. The D_2 monitoring beam originates at the top, passes from left to right, then down through the cell, and then essentially right to left horizontally. The more important labeled components are identified as follows: F_1, F_5 = Corning filters (7-69); F_2 =8944 Å pass filter (50 Å bandwidth); F_3 =8944 Å pass filter (20 Å bandwidth); F_4 =8944 Å reflect, 8521 Å pass, both at 45° incidence; F_6 =8521 Å pass filter, 8944 Å reject (30 Å bandwidth); F_7 =8521 Å pass filter, 8944 Å reject (5 Å bandwidth); *pd* is the photodetector.

tilled under a vacuum of 10^{-6} Torr or better, and an appropriate buffer gas was added. The cells were mounted in the base of the Dewar and in a detachable phenolic tube, and stabilized at the temperature of the bore, 13 °C. Measurements at higher temperatures were made by passing a current through a spiral winding of coaxial heater wire in thermal contact with a thin copper foil surrounding the cell.

C. Light sources

In order to do optical pumping at high magnetic fields, one must provide light suitable for the pumping of Zeeman sublevels widely split from the center of a normal resonance line. We employed a variation of the filtered white-light pumping scheme developed by Ensberg and zu Pulitz.[3] In that scheme, the overlap of absorption and pumping lines remains constant no matter where the absorption line is forced, and one can optically pump at any desired field with an excitation rate which is field independent. Our experiment required a white light source which was intense, stable, of low noise, small in area, and flat in intensity over the 8944-Å Cs D_1 absorption line. The best choice proved to be a 1500-W General Electric DTJ lamp, filament size about 4.5 cm², operated at 120 V dc. For the weaker detection beam we used a General Electric DHT 1200-W lamp powered by a current regulated power supply producing 7 A at about 40 V. This arrangement virtually eliminated slow dc level drifts, and reduced ac ripple on the detection beam to less than $1:10^5$.

D. Pumping beam

The light from the pumping lamp was collimated by two large (12.5-cm-diam) Pyrex condensing lenses, with an effective focal length of 10 cm. The light then passed through a water filter, a Corning 7-69 glass absorption filter, and two D_1 pass interference filters, one with 50-Å bandwidth, and one with 20-Å bandwidth, each with $D_2 : D_1$ rejection ratios of about 10^{-4}.

Following the filter train, the pumping beam was focused through a mechanical shutter which was used to generate pumping and relaxation transients. For most of the work we used a large diameter rotating wheel, as shown in Fig. 1. The wheel was separated from a variable speed motor by a long pulley to help isolate the optical detectors from the motor's mechanical and electrical noise. Shutter risetimes for the wheel ranged from 2 to 10 msec, depending upon "open time."

The horizontal pumping beam, by now filtered and chopped, was reflected vertically through a circular polarizer into the magnet bore by a specially constructed broad band interference filter which at 45° incidence reflected D_1 light, but transmitted D_2 light. The circularly polarized D_1 pumping beam then passed through the Cs vapor cell, and exited through the top of the helium Dewar.

E. Generation and detection of transient signals

We induced pumping and relaxation transients by switching the pumping beam on and off, a modifi-

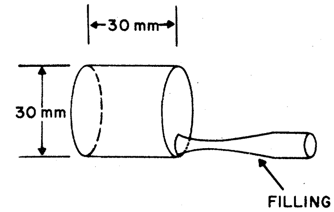

FIG. 2. Schematic of the cylindrical optical pumping cells. Cs was distilled through the filling tube, an appropriate buffer gas was added and the cell was sealed off as close to the window surface as possible. The inner dimensions of the cells were approximately 26-mm diam, 25-mm length.

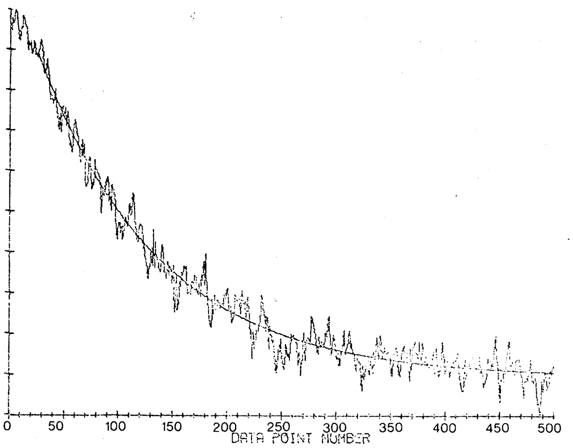

FIG. 3. Typical relaxation transient together with the best single exponential fit to the data. The data represents 2^{13} sweeps of the experimental signal produced by the relaxation of Cs in 50 Torr Ne at 13 °C and 40 kG. The sweep time is 100 msec, full scale. The shutter took about 5 msec to close at this sweep rate. The relaxation time constant for this curve was determined to be 22.0 msec.

cation of Franzen's technique for studying collisional relaxation.[2] Opening the shutter, allowed the Cs vapor to acquire a net $\langle S_z \rangle_g$ through optical pumping. Closing the shutter allowed $\langle S_z \rangle_g$ to relax back to its thermal equilibrium value. Changes in $\langle S_z \rangle_g$ were reflected in changes in the degree of absorption of circularly polarized light.

We were not able to observe transients directly on the chopped pumping beam. Changes in absorption of the 20-Å-wide pumping light corresponding to the transition of the vapor from the pumped to the unpumped state, amounted to at most a few parts in 10^5. Erratic overshoot-undershoot response of photodetectors to step function changes in light intensity are often as much as a few parts in 10^2, masking the small optical pumping signal. We therefore monitored $\langle S_z \rangle_g$ with a separate unchopped white-light detection beam passing through the cell in the direction opposite to that of the pumping beam. The detection beam incident upon the cell was unpolarized and of equal intensity in both the D_1 and D_2 lines. Nonzero $\langle S_z \rangle_g$ within the cell resulted in differential absorption of σ^+ and σ^- components, proportional to $\langle S_z \rangle_g$. The beam then passed through the pumping polarizer, which in this reverse direction of propagation acted as an analyzer, passing one sense of circular polarization only. The D_2 component of the detection beam continued through the pumping beam reflector, was reflected by several mirrors, and finally was filtered down to 5-Å bandwidth around 8521 Å. The elaborate filtering of both the chopped D_1 pumping beam and the unchopped D_2 detection beam provided better than 10^6 rejection of the pumping light by the detection system.

The detection beam finally fell upon an unbiased Schottkey-barrier photodiode.[4] The output of the photodiode, typically a few mV across 10^6 Ω, was fed to one input of a PAR 113 dc differential preamplifier, and a dc balancing voltage was applied to the other input. The difference signal was amplified, and fed to the input of a Hewlett-Packard 5480-A signal-averager system. The sweep of the signal averager was synchronously triggered with the shutter at typical repetition rates of 1–10 Hz. Between 2^{10} and 2^{14} sweeps generally were made. The averaged transient signal was recorded on punched paper tape for later computer analysis. In Fig. 3, we provide an example of a typical relaxation transient, together with the best single exponential fit to the data. The data represent 2^{13} sweeps of the experimental signal produced by the relaxation of Cs in 50-Torr Ne at 13 °C and at 40 kG. In order to ensure that our evaluations of the relaxation transients would not be perturbed by shutter risetime effects, we excluded the first few msec of data from our fits.

III. EXPERIMENTAL RESULTS

A. Relaxation measurements at 40 kG

In Figs. 4–6, we summarize measurements of the relaxation rates of Cs in He, Ne, and Ar buffer gases at 40 kG. Between three to five measurements were made at each pressure point. All data have been corrected for the contribution to the relaxation rate due to the depolarizing effect of the detection beam.[5] The 40-kG field is sufficiently

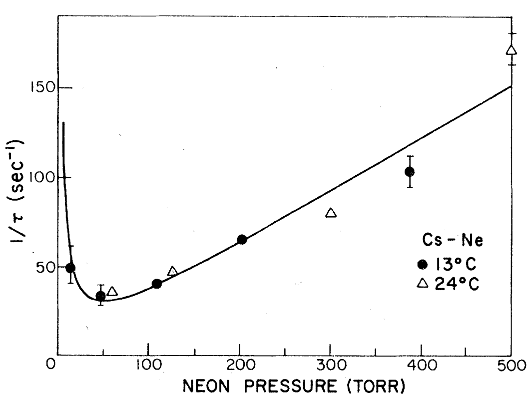

FIG. 4. Rate of relaxation of $\langle S_z \rangle_g$ of Cs as a function of Ne pressure. The magnetic field was 40 kG. The solid curve is fit of Eq. (1) to the 13 °C data, yielding $A = 751$ sec^{-1} Torr and $B = 0.30$ sec^{-1} Torr^{-1}.

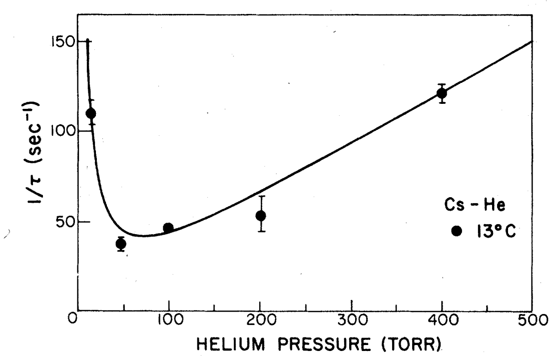

FIG. 5. Same as Fig. 4, but with He as the buffer gas. The fitted parameters are $A = 1500\ \mathrm{sec^{-1}\ Torr}$ and $B = 0.30\ \mathrm{sec^{-1}\ Torr^{-1}}$.

strong to ensure the decoupling of the electronic and nuclear spins of the Cs atom. The measured relaxation rates therefore should be unaffected by the nuclear-spin dynamics customarily encountered at low magnetic fields. Contributions to relaxation rates from the formation of bound or quasi-bound Van der Waals molecules should be negligible: At 40 kG, the angular electronic Zeeman frequency far exceeds the inverse of the characteristic time expected for such interactions. In view of these considerations, we expect that the high-field relaxation rates should depend upon buffer-gas pressure (p) in the following way:

$$(\tau)^{-1} = A/p + Bp, \tag{1}$$

where A/p is the relaxation rate at the walls of the cell, and Bp is the relaxation rate due to sudden binary collisions of Cs atoms with buffer gas atoms.[2] It has been shown that when fits of Eq. (1) are made to experimental data, that A and B in fact represent the following[6]:

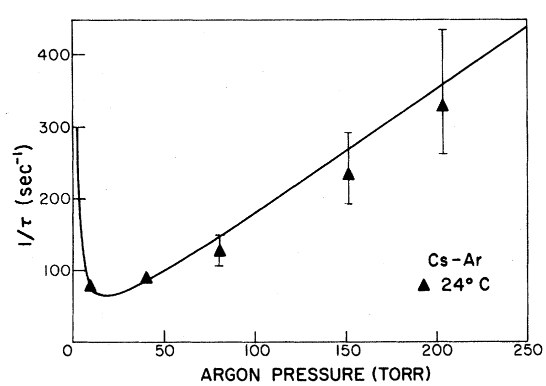

FIG. 6. Same as Fig. 4, but with Ar as the buffer gas, and with the temperatures at 24°C. The fitted parameters are: $A = 593\ \mathrm{sec^{-1}\ Torr}$ and $B = 1.77\ \mathrm{sec^{-1}\ Torr^{-1}}$.

TABLE I. Parameters of fits of Eq. (1) to data in Figs. 4–6.

	A ($\mathrm{sec^{-1}\ Torr}$)	B ($\mathrm{sec^{-1}\ Torr^{-1}}$)
Cs-He	1500	0.30
Ns-Ne	751	0.30
Cs-Ar	593	1.77

$$A \cong 1.034[(\pi/L)^2 + (2.405/r)^2] D_0 p_0, \tag{2a}$$

$$B \cong 1.074 n_0 \sigma v_{\mathrm{rel}} / p_0, \tag{2b}$$

where L and r are the length and radius of the cell, D_0 is the diffusion coefficient at atmospheric pressure (p_0), n_0 is Loschmidt's number, σ is the nuclear-spin-independent cross section for the relaxation of $\langle S_z\rangle_g$ in alkali-metal-atom–noble-gas-atom sudden binary collisions, and v_{rel} is the mean relative velocity of alkali-metal-atoms and buffer-gas atoms.

Least-square fits of Eq. (1) to the Cs-He, Cs-Ne, and Cs-Ar data are indicated by the solid curves in Figs. 4–6. The evaluated parameters A and B are listed in Table I.

B. Relaxation measurements as a function of magnetic field

In Fig. 7, we display measurements of relaxation rates for Cs in moderately high pressures of Ne. Collisions of Cs atoms with buffer-gas atoms should be the dominant relaxation mechanism in both cells. No dependence of relaxation rate upon magnetic field is evident in either case. In Table II, we summa-

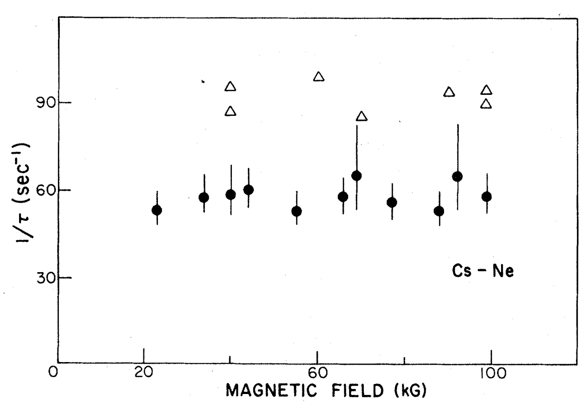

FIG. 7. Rate of relaxation of $\langle S_z\rangle_g$ of Cs vs magnetic field for a cell containing 300 Torr of Ne at 24°C (circles), and for a cell containing 138 Torr of Ne at 50°C (triangles). Each triangle represents a single measurement; each circle represents the average of two to four measurements.

TABLE II. Magnetic field dependence of the relaxation rates of $\langle S_z \rangle_g$ of Cs in 150-Torr Ar and 207-Torr He, for which buffer-gas-atom–Cs-atom collisional relaxation is dominant, and in 15-Torr He and 5-Torr Ne for which wall relaxation is dominant.

Buffer gas (p, Torr)	$(1/\tau)$ (sec^{-1}) (at 40 Kg)	$(1/\tau)$ (sec^{-1})
Ar (150)	232 ±48	244 ± 18 (97 Kg)
He (207)	55 ±10	47 ± 6 (90 Kg)
He (15)	112 ±8	92 ± 8 (90 Kg)
Ne (5)	204 ±37	152 ± 7 (97 Kg)

rize the results of additional measurements made on a 150-Torr Ar cell, and a 207-Torr He cell, which also display no significant dependence upon magnetic field. Relaxation rates for cells containing 15 Torr of He and 5 Torr of Ne, on the other hand, appear diminished at very high magnetic fields (see Table II). We shall use these various results in the following section to gain information on correlation times for several collisional interactions.

IV. ANALYSES AND DISCUSSION

A. Diffusion coefficients and wall relaxation

Equation (2) and the results in Table I yield the values of the diffusion coefficients for Cs in He, Ne, and Ar listed in the first column of Table II. In columns 2–5, we compare these determinations with earlier measurements made at low magnetic field. We include those made by Franz and Sooriamoorthi (FS) from measurements of the relaxation of $\langle S_z \rangle_g$ in low magnetic fields[7]; those made by Beverini *et al.* from measurements of the relaxation of $\langle S \cdot I \rangle_g$ in intermediate magnetic fields[8]; those made by Kanda and Minemoto from measurements of the relaxation of F_+[9]; and those made by Mioduszewska-Grochowska, Skubiszak, and Rosinski on the relaxation of $\langle S_x \rangle$.[10] For ease of comparison, all values have been extrapolated to 273 °K, assuming $T^{3/2}$ scaling.

The only significant difference in the various determinations of the D_0's lies in that for Cs-He, where the value measured by Beverini *et al.* falls 40% below our value and 30% below that measured by FS. The data from which Beverini *et al.* evaluated D_0 span a rather narrow range of relaxation rates compared to the other evaluations, however. We believe that the value of D_0 for Cs-He determined by FS, which is about 15% less than our high-field value, is the most reliable of the three determinations.

The overall agreement of the values of the diffusion coefficients determined by the various methods, implies that the relaxation rates of Cs on glass walls are the same for all observables. This, in turn, lends very strong support to the "uniform" model of relaxation of alkali-metal atoms on optical pumping cell walls.[2] In the "uniform" model, it is assumed that any Zeeman sublevel of the $^2S_{1/2}$ alkali-metal-atom ground state can be reached with equal probability in a single relaxation event. One consequence of the model is that wall relaxation rates must be nuclear spin independent: The same wall relaxation rates must exist for the observables $\langle S_z \rangle_g$, $\langle I_z \rangle_g$, and $\langle S \cdot I \rangle_g$ at low magnetic field as for $\langle S_z \rangle_g$ at moderately high magnetic fields. It is exactly this situation which is borne out by the measurements compared in Table II. The results contrast sharply with the quite different situation of relaxation on glass walls coated with long-chain hydrocarbons.[11]

B. Relaxation in sudden binary collisions of Cs atoms with buffer-gas atoms

It is generally believed that electronic spin relaxation in sudden binary collisions of alkali-metal atoms with noble-gas atoms occurs through a spin-orbit interaction $\gamma(\vec{S} \cdot \vec{N})$, where $\vec{N}$ is the translational angular momentum of the colliding pair.[12–15] An approximate description of the effect of the $\gamma(\vec{S} \cdot \vec{N})$ interaction can be obtained by assuming that through collisions the alkali-metal-atom is subjected to a randomly occurring, randomly oriented magnetic field of strength h_1, and of average duration τ_{e1}. The re-

TABLE III. Comparison of the present determination of D_0 (high H_0) with those made by other methods. All values have been extrapolated to 273 °K assuming $D = D_0(T/273)^{3/2}$.

D_0(cm^2/sec)	$\langle S_z \rangle$ (high H_0)	$\langle S_z \rangle$ (low H_0)[a]	$\langle \vec{S} \cdot \vec{I} \rangle$[b]	$T_2(F_+)$[c]	$\langle S_x \rangle$[d]
Cs-Ar	0.13	0.11	0.13	0.12	0.10
Cs-Ne	0.17	0.17	0.15	0.16	0.20
Cs-He	0.34	0.29	0.20	···	0.33

[a]Reference 7.
[b]Reference 8.
[c]Reference 9.
[d]Reference 10.

TABLE IV. Rates of relaxation in alkali-metal-atom–buffer-gas-atom sudden binary collisions as predicted from low-field $\gamma(\vec{S}\cdot\vec{N})$ cross sections (column 1),[7] compared to relaxation rates measured at 40 Kg (column 2). The differences, column 3, are due to the contribution of the $\delta a(\vec{S}\cdot\vec{I})$ interaction at high magnetic field. p is buffer-gas pressure in Torr.

	R predicted ($\sec^{-1}$)	R measured ($\sec^{-1}$)	Difference ($\sec^{-1}$)
Cs-He	$0.10p$	$0.28p$	$0.18p$
Cs-Ne	$0.08p$	$0.28p$	$0.20p$
Cs-Ar	$1.65p$	$1.62p$	$0.03p$

laxation rate R_1 due to this perturbation then can be calculated to be[14,16]

$$R_1 = \frac{2}{3}\,\frac{(\gamma_e h_1)^2 \tau_{e1}^2 (n_0 \sigma_g v_{\rm rel}\; p/p_0)}{1+\omega_0^2 \tau_{e1}^2}, \tag{3}$$

where ω_0 is the resonant frequency for the electronic-spin polarization, γ_e is the electronic gyromagnetic ratio, n_0 is Loschmidt's number, $v_{\rm rel}$ is the mean relative velocity of alkali-metal atoms and buffer-gas atoms, p is the buffer-gas pressure, and p_0 is atmospheric pressure. σ_g is an average cross section describing the range of the $\gamma(\vec{S}\cdot\vec{N})$ interaction, and can be taken as roughly equivalent to the geometric (hard-sphere) cross section.[13] More often, Eq. (3) is represented empirically in terms of a cross section for relaxation,

$$R_1 = n_0 \sigma v_{\rm rel}\; p/p_0 , \tag{4}$$

where σ is the nuclear-spin-independent cross section for relaxation via the $\gamma(\vec{S}\cdot\vec{N})$ interaction.

The best determinations of σ for Cs at present are those of Franz and Sooriamoorthi, who obtained $\sigma(\text{Cs-He}) = 2.43\times10^{-23}$ cm^2, $\sigma(\text{Cs-Ne}) = 4.08\times10^{-23}$ cm^2, and $\sigma(\text{Cs-Ar}) = 108\times10^{-23}$ cm^2, all with estimated uncertainties of $\pm10\%$.[7,17] Using these cross sections, one can calculate from Eq. (4) the relaxation rates that would be expected at high magnetic fields. The measured and predicted rates for Cs-Ar turn out to be almost equal, as can be seen in Table IV. The measured high-field relaxation rates for Cs-He and Cs-Ne, however, turn out to be several times larger than those predicted from the low-field data. To dramatize the effect, in Fig. 8, we compare the high-field relaxation rates for Cs-Ne, with the wall relaxation rates subtracted out from the low-field projections. The puzzle is compounded by the fact that the relaxation rates for Cs-He and Cs-Ne do not change with magnetic field in the region 20–100 kG (see Fig. 7 and Table II). The possibility that the enhancement of the high-field relaxation rate could be due to a mechanism such as a magnetic field induced small admixture of non-S states into the ground state appears to be ruled out, since such an effect should exhibit a continuous dependence upon strength of magnetic field, which is not observed.

We now shall show that the enhanced relaxation measured at high magnetic field arises from the modification of the Cs hyperfine interaction $\delta a(\vec{S}\cdot\vec{I})$ induced in sudden binary collisions of Cs atoms with noble-gas atoms, the same interaction that is responsible for the well-known buffer-gas pressure shift of alkali-metal-atom hyperfine frequencies.[1] Abragam has considered the influence of the $\delta a(\vec{S}\cdot\vec{I})$ interaction on a system of unbound, decoupled spins in a liquid.[18] The extension to the case of alkali-metal atoms suffering collisions in high magnetic fields is straightforward.[19] We obtain

$$R_2 = \frac{2}{3}\,\frac{\Omega^2 \tau_{e2}^2 I(I+1)(n_0 \sigma_c v_{\rm rel}\; p/p_0)}{1+\omega_0^2 \tau_{e2}^2}. \tag{5}$$

Ω is 2π times the average shift in a ($\langle\delta a\rangle$) per collision, σ_c is the average collisional cross section associated with this interaction, and τ_{e2} is the average duration of the $\delta a(\vec{S}\cdot\vec{I})$ interaction. $\langle\delta a\rangle$ can be calculated from low-field measurements of buffer-gas pressure shifts

$$\langle\delta a\rangle \cong \frac{(\delta\nu)p}{I+\frac{1}{2}}\;\frac{(n_0\sigma_c v_{\rm rel}\; p/p_0)^{-1}}{\tau_{e2}}, \tag{6}$$

where $\delta\nu$ is the measured pressure shift in Hz/Torr. If we assume that parameters such as $\delta\nu$, τ_{e2}, and σ_c are not themselves functions of magnetic field, we finally obtain

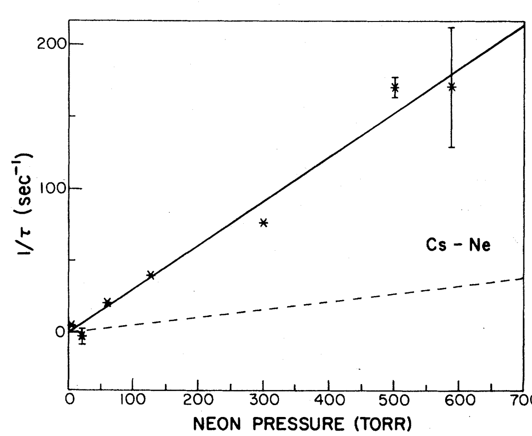

FIG. 8. Comparison of the relaxation rates measured in Cs-Ne at 40 kG, 24 °C, with predictions based on the low-field $\gamma(\vec{S}\cdot\vec{N})$ cross sections alone (dashed line). The wall relaxation rates have been subtracted from the data. Addition of contributions from the $\delta a(\vec{S}\cdot\vec{I})$ interaction yields the fitted line passing through the data.

$$R_2 = \frac{8\pi^2}{3}\frac{(\delta\nu)^2}{(I+\frac{1}{2})^2}\frac{(n_0\sigma_c v_{\rm rel}/p_0)^{-1}}{(1+\omega_0^2\tau_{e2}^2)}p(I)(I+1), \qquad (7)$$

which should fully describe the contribution of the $\delta a(\vec{S}\cdot\vec{I})$ interaction to $\langle S_z\rangle_g$ relaxation at high magnetic field.

We now shall calculate the expected contributions of R_2 for Cs buffer-gas collisions at 40 kG. Since we observed no dependence of Cs buffer-gas relaxation rates upon magnetic field in the range 20–100 kG, we can conclude that at 40 kG the condition $\omega_0^2\tau_{e2}^2 \ll 1$ still holds, thus effectively eliminating the dependence of R_2 upon τ_{e2} at that field. The pressure shifts, $\delta\nu$, have been measured independently,[20–22] and are listed in Table V. The values of all parameters in Eq. (7) therefore are fixed, with the exception of σ_c. Even σ_c is roughly known, however: It would be difficult to understand why σ_c for Cs-He and Cs-Ne should be substantially greater than πR_m^2, where R_m is the internuclear separation at the minimum of the alkali-atom–noble-gas-atom Van der Waals potential. It is only for separations less than R_m that strong repulsion occurs, and with it significant shifts of the Cs electron density at the Cs nucleus giving rise to positive hyperfine frequency shifts.[23] In Column 2 of Table V, we list the values of σ_c for Cs-He and Cs-Ne that are required in Eq. (7) to produce the values of R_2 that have been measured: The results indeed turn out to be roughly of the size πR_m^2. We conclude that the sum of contributions from the $\gamma(\vec{S}\cdot\vec{N})$ and $\delta a(\vec{S}\cdot\vec{I})$ interactions fully accounts for the Cs buffer-gas relaxation observed at high magnetic fields.

We now shall explain why essentially equal relaxation rates are measured at high and low magnetic fields in the case of Cs-Ar. In accord with the discussion of the preceding paragraph, we assume that σ_c(Cs-Ar) $\cong 1\times10^{-14}$ cm^2. Eq. (7) then predicts that relaxation due to the $\delta a(\vec{S}\cdot\vec{I})$ interaction for Cs-Ar should be approximately $0.02p$ sec^{-1}, a contribution which is almost negligible compared to that arising from the $\gamma(\vec{S}\cdot\vec{N})$ interaction for Cs-Ar, about $1.62p$ sec^{-1}. Since the contribution from $\gamma(\vec{S}\cdot\vec{N})$ should be the same at low and moderately high fields, and since $\delta a(\vec{S}\cdot\vec{I})$ makes a negligible contribution in both cases, we expect no significant change of the overall relaxation rate in Cs-Ar in moving from low to high magnetic field, in accord with the experimental result.

We still must explain how the $\delta a(\vec{S}\cdot\vec{I})$ interaction can make large contributions to Cs-He and Cs-Ne relaxation in high magnetic fields, but not in low magnetic fields. The answer is straightforward: in the presence of very small magnetic fields the Hamiltonian is diagonal in (F, m_F) and population changing transitions cannot be induced. The more general case of collisions occurring in the presence of intermediate coupling between $\vec{S}$ and $\vec{I}$ has been treated by Bouchiat[19] and by Volk.[24,25] The result in that case is that Eq. (7) is multiplied by approximately the factor $(\omega_s/\Delta W)^2$, where ω_s is the angular electronic Zeeman frequency in the external magnetic field, and ΔW is the angular hyperfine frequency. At 1 G $\omega_s/\Delta W$ is of the order of 10^{-3}, implying a negligible contribution of the $\delta a(\vec{S}\cdot\vec{I})$ interaction to relaxation rates at low magnetic field. At 40 kG, however, $\omega_s/\Delta W \approx 12$, and the straightforward high-field results, Eq. (7) unmodified by the $(\omega_s/\Delta W)^2$ factor, apply. The transition region in which a reasonably strong dependence of the contribution from the $\delta a(\vec{S}\cdot\vec{I})$ interaction upon magnetic field should be measurable, lies in the range where $\omega_s/\Delta W \approx 1$, that is, at a magnetic field of the order of 3 kG. Analyses of experimental results in that region, however, are complicated by the fact neither low- nor high-field approximations to normal nuclear-spin mechanics apply.

TABLE V. Measured pressure shifts $\delta\nu$ for Cs^{133}, and evaluated cross sections σ_c for the $\delta a(\vec{S}\cdot\vec{I})$ interaction. The values of Ref. 20 were used in our calculations.

	$\delta\nu$(Hz/Torr)	$\sigma_c(10^{-14}$ cm$^2)$
Cs-He	1050 [a]	
	1600 [b,c]	1.3
	1200 [b,c]	
Cs-Ne	580 [a]	
	650 [b,c]	0.73
Cs-Ar	−190 [a]	
	−250 [b,c]	Not measured
	−212 [b,c]	

[a]Reference 20. [c]Reference 22.
[b]Reference 21.

C. Average durations of collisional interactions

The measurements of relaxation rates as a function of magnetic field yield information on the magnitudes of the average durations for the various collisional interactions. Estimating conservatively that a 20% change in relaxation rate should have been discernable in our experiment, we can set limits on τ_e in cases where no appreciable change in relaxation rate was observed. In the 150-Torr Cs-Ar cell, for example, (see Table II) by applying the criterion that R(100 kG) ≥ 0.8 $R(\approx 0$ kG), we obtain τ_e(Cs-Ar) $\leq 2.8\times10^{-13}$ sec. From the discussion in Sec. IV B, above, we know that virtually all of the relaxation in this cell arises from the $\gamma(\vec{S}\cdot\vec{N})$ interaction in Cs-Ar collisions. We therefore conclude that

$$\tau(\text{Cs-Ar})\,(\vec{S}\cdot\vec{N}) \leq 2.8\times 10^{-13}\ \text{sec}.$$

We note that if τ_e scales as the relative velocity of the colliding pair, this implies that

$$\tau(\text{Cs-He})\,(\vec{S}\cdot\vec{N}) \leq 1\times 10^{-13}\ \text{sec}.$$

The 138- and 300-Torr Cs-Ne cells also display no dependence of R upon H_0 (see Fig. 7). In this case, however, R is made up of contributions from both $\gamma(\vec{S}\cdot\vec{N})$ and $\delta a(\vec{S}\cdot\vec{I})$ interactions: $\delta a(\vec{S}\cdot\vec{I})$ in fact dominates. Using the data in Table II, and the 20% criterion, we estimate that

$$R_2(\text{Cs-Ne})(100\,\text{kG}) \geq 0.7\ R_2(\text{Cs-Ne});\ (\approx 0\ \text{kG}),$$

from which we obtain

$$\tau_e(\text{Cs-Ne})(\delta a(\vec{S}\cdot\vec{I})) \leq 3.6\times 10^{-13}\ \text{sec}.$$

Again, if τ_e scales roughly as v_{rel}, this implies that

$$\tau_e\,(\text{Cs-He})(\delta a(\vec{S}\cdot\vec{I})) \leq 1.7\times 10^{-13}\ \text{sec}.$$

In Table II, we presented data on two low-pressure cells, 15-Torr He, and 5-Torr Ne, in which wall relaxation is dominant (96% dominant in the He cell, 99% in the Ne cell). At magnetic fields approaching 100 kG, both cells had relaxation times which were significantly longer than those which were measured at 40 kG, 22% longer in the 15-Torr He cell, and 35% longer in the 5-Torr Ne cell. If we assume that the relaxation rate, R' measured at low buffer-gas pressures is due to weak magnetic relaxation of Cs atoms on cell surfaces rather than to escape and replacement of atoms from the cell, then R' should have a dependence upon H_0 similar to that in Eq. (3), that is,

$$R' \propto [1+(\gamma_e H_0)^2]^{-1}.$$

Utilizing the data in Table II, we thus obtain

$$\tau_e(\text{walls}) \geq 3.5\times 10^{-13}\ \text{sec}$$

for the Cs-He cell, and

$$\tau_e\,(\text{walls}) \geq 4.0\times 10^{-13}\ \text{sec}$$

for the Cs-Ne cell. These two results are not significantly different, as should be the case: τ_e for alkali-wall collisions should not be dependent upon the type of buffer gas present in the cell. We determine, therefore, that

$$\tau_e(\text{walls}) \geq 4\times 10^{-13}\ \text{sec}$$

for Cs relaxation in collisions with alkali-coated-Pyrex glass surfaces. The inequality sign is maintained because we cannot rule out the possibility that a portion of the apparent relaxation rate may be due to escape and replacement: Our value of 4×10^{-13} sec therefore is a *lower* limit for τ_e (walls), under the ambient experimental conditions.

D. Comparison of measurements at high and low magnetic fields

Comparisons of rates for alkali spin relaxation measured by different methods sometimes have been confused by the unsuspected presence of anomalous contributions to relaxation from the formation of bound and quasibound Van der Waals molecules, or by the incorrect treatment of nuclear-spin effects. Most discrepancies and disagreements between experiments disappear, however, when these effects are properly treated. It is an important consistency check, therefore, to measure and evaluate some low-field relaxation rates in the actual cells used in the present experiment. In column 1 of Table VI, we record measurements of the difference Z_2-Z_1 of the two rate constants which make up the low magnetic field optical pumping transient of $\langle S_z\rangle_g$ for several cells for which high-field measurements have already been given in Figs. 4 and 5. The data were obtained at 23 °C, 0.20 G, in the white light optical pumping rig described by Sooriamoorthi.[26] At low field, Z_2-Z_1 is given approximately by

$$Z_2-Z_1 \approx \tfrac{31}{32}R + \tfrac{20}{32}Rs + R^*, \tag{8}$$

where R is the relaxation rate in alkali-metal-atom–buffer-gas-atom sudden binary collisions, R_s is the spin exchange rate, and R^* is the anomalous (molecular) relaxation rate.[27] We recall from our earlier discussion that at low fields, R should contain contributions only from the $\gamma(\vec{S}\cdot\vec{N})$ inter-

TABLE VI. Measured and predicted values of relaxation rates at low and high magnetic fields in typical optical pumping cells used in this experiment. The large enhancements of the measured high-field relaxation rates are due to contributions from the $\delta a(\vec{S}\cdot\vec{I})$ interaction (see text).

Buffer gas (Torr)	Measured (Z_2-Z_1) (low H_0, sec^{-1})	Predicted (Z_2-Z_1) (low H_0, sec^{-1})	Predicted $(1/\tau)$ (high H_0, sec^{-1})	Measured $(1/\tau)$ (high H_0, sec^{-1})
500 Ne	65	63	42	172
300 Ne	48	47	27	80
412 He	65	61	45	120

action. Using the results of Ref. 7, we predict the values for (Z_2-Z_1) which are listed in column 2 of Table VI: these agree very well with the measurements in column 1. For sake of completeness, in column 3 we list the relaxation rates that would be measured at high field, assuming that only the $\gamma(\vec{S}\cdot\vec{N})$ interaction were to contribute in Cs-He and Cs-Ne collisions. The measured high-field relaxation rates are listed in column 4. The large differences between columns 3 and 4 are due to the added contribution of the $\delta a(\vec{S}\cdot\vec{I})$ interaction, as we have explained in earlier sections.

---

*Research supported by the Air Force Office of Scientific Research, Office of Aerospace Research, under Grant No. 74-2652.

†Work performed in partial fulfillment of the requirements for the Ph.D. degree in Physics at Indiana University, Bloomington, Ind.

[1]For a review of optical pumping experiments and theory, see W. Happer, Rev. Mod. Phys. 44, 169 (1972).

[2]The experiment involves a modification of techniques devised by H. G. Dehmelt [Phys. Rev. 105, 1487 (1957)] and W. Franzen [*ibid.* 115, 850 (1959)].

[3]E. S. Ensberg and G. zu Putlitz, Phys. Rev. Lett. 22, 1349 (1969).

[4]PINIOD, available from United Detector Technology, Santa Monica, Calif.

[5]T. R. Marshall, Ph.D. thesis (Indiana University, Bloomington, Ind., 1975 (unpublished). Typical corrections were 5% of the measured relaxation rate, with no correction exceeding 10%.

[6]F. A. Franz, Phys. Rev. A 6, 1921 (1972).

[7]F. A. Franz and C. E. Sooriamoorthi, Phys. Rev. A 10, 126 (1974).

[8]N. Beverini, P. Minguzzi, and F. Strumia, Phys. Rev. A 4, 550 (1971).

[9]T. Minemoto and T. Kanda, J. Phys. Soc. Jpn. 31, 1174 (1971).

[10]B. Mioduszewska-Grochowska, W. Skobiszak, and K. Rosinski, Lett. Nuovo Cimento 9, 403 (1974).

[11]See, for example, M. A. Bouchiat, J. Phys. (Paris) 24, 379 (1963); 24, 611 (1963); and G. Moruzzi, N. Inguscio, F. Strumia, and P. Violino, Phys. Rev. A 8, 51 (1973).

[12]R. A. Bernheim, J. Chem. Phys. 36, 135 (1962).

[13]R. H. Herman, Phys. Rev. 136, A1576 (1964).

[14]C. C. Bouchiat, M. A. Bouchiat, and L. C. L. Pottier, Phys. Rev. 181, 144 (1969).

[15]C. C. Bouchiat and M. A. Bouchiat, Phys. Rev. A 2, 1274 (1970).

[16]C. P. Slichter, *Principles of Magnetic Resonance* (Harper and Row, New York, 1963), p. 141, Eq. 137.

[17]N. Beverini, P. Minguzzi, and F. Strumia, Ref. 8; and A. Sieradzan, J. Dresner, and K. Rosinski, Optics Commun. 17, 83 (1976), found values of σ(Cs-He) and σ(Cs-Ar) comparable to those reported in Ref. 7, but reported significantly different values of σ(Cs-Ne). The Cs-Ne results can be reconciled when anomalous relaxation via van-der-Waals molecular formation is properly accounted for, as we shall show in a separate publication. The high-pressure results of Minemoto and Kanda (Ref. 9), should not be significantly affected by anomalous relaxation. Their reported cross section for Cs-Ne coincides with that of Ref. 7.

[18]A. Abragam, *The Principles of Nuclear Magnetism* (Oxford, U.P., New York, 1961), pp. 305–312.

[19]See also, M. A. Bouchiat, Ph.D. thesis (University of Paris, 1964) (unpublished), p. 150.

[20]E. C. Beaty, P. L. Bender, and R. A. Chi, Phys. Rev. 112, 450 (1958).

[21]M. Arditi and T. R. Carver, Phys. Rev. 109, 1012 (1958); 112, 449 (1958); 124, 800 (1961).

[22]M. Arditi, J. Phys. Radium, 19, 873 (1958).

[23]See, for example, R. R. Freeman, E. M. Mattison, D. E. Pritchard, and D. Kleppner, Phys. Rev. Lett. 33, 397 (1974). These authors measured significant hyperfine frequency shifts in the K-Ar Van der Waals molecule.

[24]C. Volk, Ph.D. thesis (Indiana University, Bloomington, Ind., 1977) (unpublished).

[25]For a treatment of the effect of the $\delta a(\vec{S}\cdot\vec{I})$ interaction on line shift and line broadening, see, for example, F. Hartmann and F. Hartmann-Boutron, Phys. Rev. A 2, 1885 (1970).

[26]C. E. Sooriamoorthi, Ph.D. thesis (Indiana University, Bloomington, Ind., 1974) (unpublished).

[27]F. A. Franz and C. Volk, Phys. Rev. A 14, 1711 (1976).