

Foreign-Gas-Induced Cesium Hyperfine Relaxation*

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We have measured the hyperfine relaxation time of cesium vapor in the presence of foreign gases. We have therefore been able to measure the diffusion coefficient D_0 and the cross section $\sigma(\vec{s} \cdot \vec{I})$ of hyperfine relaxation due to the collisions with the foreign gas. The values of D_0 for He, Ne, Ar, and N_2 are 0.204 ± 0.04 , 0.153 ± 0.014 , 0.134 ± 0.02 , and 0.073 ± 0.015 cm^2/sec at 0°C and 760 Torr, respectively. The cross sections are 2.80 ± 0.3 , 9.27 ± 0.9 , 104 ± 10 , and 55.2 ± 4.4 in units of 10^{-23} cm^2 for He, Ne, Ar, and N_2 , respectively. We have also improved on the measurement of the spin-exchange cross section σ_{ex} obtaining the value $(2.18 \pm 0.12) \times 10^{-14}$ cm^2 . We compare the problems arising in the study of hyperfine and Zeeman relaxation and we discuss the results.

I. INTRODUCTION

After Franzen's pioneering work¹ many experimental and theoretical papers have been concerned with the study of the relaxation of optically pumped alkali vapors in the presence of foreign gases. The problem is rather complicated by several interacting phenomena. At the beginning, many of these effects were not known; therefore in many experiments not all of the necessary parameters have been controlled. Consequently only qualitative results are available from many experimental data. These experiments are essentially of two different kinds: In a first case the atoms are oriented by means of a light beam with circular polarization and one measures the relaxation of the observable $\langle S_z \rangle$, that is the orientation of the electron spin. In a second case the atoms undergo hyperfine pumping by using a filtered light whose spectrum contains a single component in the hyperfine doublet of the resonance lines. Then the observable of interest is $\langle \vec{S} \cdot \vec{I} \rangle$ which, in the ground state of an alkali atom, is linked to the difference of occupation numbers of the two hyperfine sublevels.

Since the relaxation of $\langle S_z \rangle$ is more easily measured, it has been studied for all of the alkali metals; however there are still unsolved problems in this field. The study of the hyperfine relaxation is usually more difficult because it requires a hyperfine filter for the resonance lines. However the theoretical treatment of the results is simpler and one can obtain some interesting information.

In this paper we report the results of the study of the hyperfine relaxation of cesium in the presence of different buffer gases. We have used two different experimental apparatus as described in detail in Sec. II and we have obtained a very effective hyperfine pumping for cesium vapors.² We have studied the systems Cs-He, Cs-Ne, Cs-Ar, Cs- N_2 , and for each of these pairs the results are (a) the cross-section $\sigma(\vec{s} \cdot \vec{I})$ for the process of destruction of the

hyperfine pumping and (b) the diffusion coefficient D_0 of the Cs atoms in the buffer gas.

Preliminary results for the pair Cs-Ne have already been published.³

The cross section σ_{ex} for the spin-exchange process between cesium atoms has been measured again with increased statistical accuracy: This val-

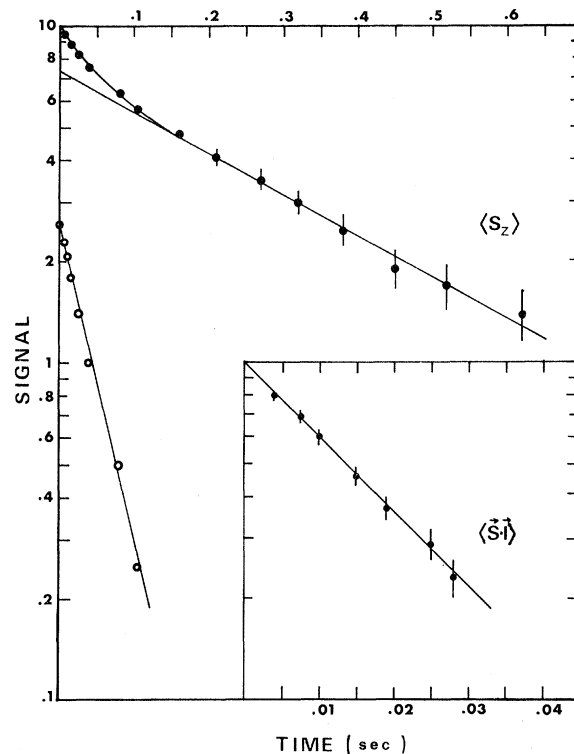


FIG. 1. Relaxation transients in a cell containing 74 Torr of Ne, recorded with Franzen's method. The hyperfine relaxation $\langle \vec{S} \cdot \vec{I} \rangle$ is a single exponential, while the Zeeman relaxation $\langle S_z \rangle$ is the sum of two exponentials within the experimental errors (the circles are the difference between the signal and a single exponential).

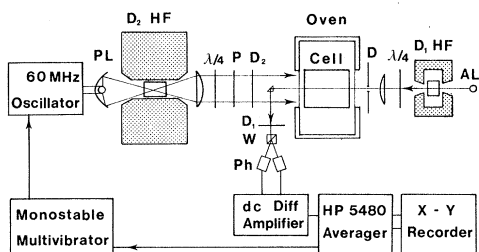


FIG. 2. Experimental apparatus for the continuous recording of the hyperfine relaxation: pumping lamp (PL), D_2 hyperfine filter (D_2 HF), linear polarizer (P), interference filters (D_2 and D_1), Wollaston prism (W); photomultiplier tube (Ph), diaphragm (D), D_1 hyperfine filter (D_1 HF), and analysing lamp (AL).

ue is the result of the measurements on all of the above-mentioned cesium-buffer-gas pairs.

II. EXPERIMENTAL APPARATUS

The measurements with He, Ne, and Ar as buffer gases have been performed with the experimental apparatus described in Refs. 2 and 4. The hyperfine pumping of cesium vapor is achieved by means of the $F = 4$ component in the D_1 resonance line. Franzen's method is used to observe the relaxation transients, and the light transmitted by the cell is monitored with a silicon solar cell in order to minimize the low-frequency transient response of the detector. In Fig. 1 we show a typical result for the hyperfine relaxation together with a transient of the Zeeman relaxation showing the characteristic two-exponential decay.

Recently a new type of cesium hyperfine filter has been set up⁵ and we have used it for the pair Cs- N_2 in a more refined experimental apparatus (Fig. 2). A high-efficiency hyperfine filter for the D_2 line is used on the pumping light beam. A second light beam passes through the cell in the opposite direction along its axis. This detection light is prepared by means of a low-field magnetic filter.⁵ After the $\frac{1}{4}\lambda$ plate the hyperfine components $F = 3$ and $F = 4$ of the D_1 line have orthogonal linear polarization states. Therefore, they can be split into two beams by means of a Wollaston prism and they are detected separately with two different photomultipliers. When the cesium vapor undergoes hyperfine pumping, because of the different absorption of the two hyperfine components, the two photomultipliers produce signals of opposite sign which are fed into a dc coupled differential amplifier. So the output signal is doubled and the noise from the lamp intensity fluctuations is greatly reduced. The stray pumping light (D_2 line) reflected or diffused by the cell walls is eliminated by means of an interference filter for the D_1 line, placed before the Wollaston prism.

As the relaxation transient can be recorded con-

tinuously when the pumping lamp is turned off (Fig. 3), it is possible to use a digital averager in order to increase the signal-to-noise ratio. The starting pulse for the measurement cycle is a gate pulse from the averager. This signal triggers a monostable multivibrator that turns off the pumping light supply; the switching time is less than 1 msec. After a preset time—determined by the multivibrator time constant—the lamp is turned on again. If necessary, up to a few thousands of transients can be averaged without any trouble. With this method we were able to measure relaxation times shorter than 10^{-2} sec with good precision.

The absorption cells of cylindrical shape are made out of Pyrex glass with an inside diameter of 5.5 cm and a length of about 7.5 cm; they are placed in a Perspex box in which thermostated water circulates. The precision in the temperature reading is about ± 0.1 deg and the stability in the temperature of the cell is definitely better than this. The temperature range for the measurements of the hyperfine relaxation time lies between 5 and 32°C .

III. RESULTS

From the relaxation time τ , which is actually observed, one can deduce the relaxation rate $1/\tau_0$ for collisions against the buffer gas and the walls by extrapolation to zero alkali density.

One can, in fact, assume that

$$1/\tau = 1/\tau_0 + 1/\tau_{\text{ex}}, \quad (1)$$

where $1/\tau_{\text{ex}}$ accounts for the spin-exchange relaxation. In Fig. 4 we show a typical extrapolation line and, for comparison purposes, also the density dependence of the relaxation of $\langle S_z \rangle$ when measured in similar conditions. It is worth noting that, while the relaxation rate $1/\tau$ for $\langle \tilde{S} \cdot \tilde{I} \rangle$ is rapidly changing, $1/\tau$ for $\langle S_z \rangle$ is almost constant. This result,

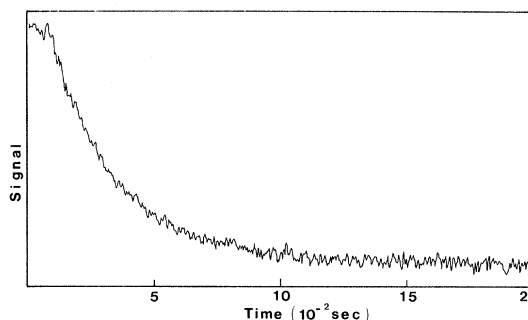


FIG. 3. Experimental recording of the hyperfine relaxation of cesium with 22.4 Torr of N_2 as a buffer gas. The temperature of the cell is 17.3°C and the total sweep time 200 msec.

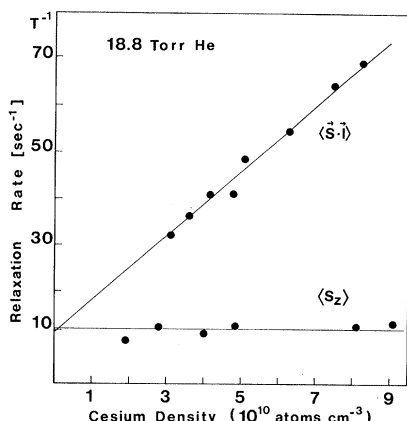


FIG. 4. Relaxation rate for $\langle \vec{S} \cdot \vec{I} \rangle$ and $\langle S_z \rangle$ vs cesium density.

as already discussed in Ref. 2, proves that the temperature dependence of the diffusion process is negligible in the pressure and temperature range we used.

A. Spin-Exchange Cross Section

In the extrapolation of the measured relaxation time to zero alkali density, one obtains two kinds of data: the intercept $1/\tau_0$, from which $\sigma_{\langle \vec{S} \cdot \vec{I} \rangle}$ and D_0 are determined, and the slope $\partial(1/\tau)/\partial N$, which is linked to the cross section σ_{ex} for spin exchange Cs-Cs. Therefore for every buffer gas and every pressure value one obtains a measurement of σ_{ex} : The result we obtained as a mean of 21 measurements is

$$\sigma_{\text{ex}} = (2.18 \pm 0.12) \times 10^{-14} \text{ cm}^2. \quad (2)$$

We have assumed that the temperature dependence of the diffusion process is negligible in comparison with the stronger temperature dependence caused by spin-exchange collisions. This assumption is in agreement with experimental data always showing straight lines for the curves of the relaxation rate versus Cs density (except in two less reliable cases which have been discarded).

The quoted error is partly statistical and includes an estimate of possible systematic errors from the Cs density formula as discussed in Sec. IV.

B. Density Problems

Both σ_{ex} and τ_0 (i. e., D_0 and $\sigma_{\langle \vec{S} \cdot \vec{I} \rangle}$) depend on which function is assumed in order to compute cesium-vapor density at the different temperatures. All the above quoted results have been obtained with use of the Taylor-Langmuir⁶ formula which has been experimentally checked by Rozwadowski and Lipworth⁷ in the low-temperature interval.

Important errors are possible if the cesium pressure in the cell is different from the saturated vapor

pressure at the temperature of operation. In order to minimize these errors we have used a large amount of cesium metal in the cells and we have carefully controlled the operating conditions. A further test comes from the comparison between the zero-density extrapolation for the relaxation probabilities of $\langle S_z \rangle$ and $\langle \vec{S} \cdot \vec{I} \rangle$. A correct formula for cesium density must, in fact, give equal limit values for the two probabilities at low buffer-gas pressure (see Fig. 4) because the diffusion process is the same in the two cases and the relaxation here is almost completely caused by wall collisions.

The error in the density of saturated vapor computed for an exact temperature value by Taylor-Langmuir formula is about 3% (see Ref. 7); a further error in the density is caused by the temperature indetermination and it is of the order of 1.5%. The effect of these fluctuations on the measurement of σ_{ex} is of the order of 3.5% as we have tested by changing the density curve by this amount.

C. Diffusion and Hyperfine Relaxation

The extrapolated relaxation times τ_0 depend on the two parameters D_0 and $\sigma_{\langle \vec{S} \cdot \vec{I} \rangle}$. A theoretical expression for the relaxation signal $\Delta(t)$ versus time is obtained from the solution of a diffusion equation^{1,8} with boundary conditions according to the geometry of the system. The result for completely disorienting wall collisions is the following function^{3,8}:

$$\Delta(t) = \Delta_0 \sum_{i\nu} B_{i\nu} e^{-t/\tau_{i\nu}}, \quad (3)$$

where $\tau_{i\nu}$ are the "relaxation times" of the different diffusion modes and the weights $B_{i\nu}$ depend mainly on the geometry of the system as discussed below. Detailed formulas are given in the Appendix. The theoretical relaxation time t_r is defined by the equation

$$\Delta(t_r) = (1/e)\Delta(t=0). \quad (4)$$

TABLE I. Diffusion coefficient D_0 (in units of cm^2/sec) and hyperfine cross section $\sigma_{\langle \vec{S} \cdot \vec{I} \rangle}$ (in units of 10^{-23} cm^2) for all of the buffer gases studied in this paper. For comparison purpose we report also the D_0 value obtained with Zeeman relaxation studies by different authors and the theoretical results from gas kinetic data.

Gas	$\sigma_{\langle \vec{S} \cdot \vec{I} \rangle}$	D_0	D_0	D_0	D_0
		This paper	Ref. 14	Ref. 15	Theoret ^a
He	2.80 ± 0.30	0.204 ± 0.04		0.37	0.29
Ne	9.27 ± 0.90	0.153 ± 0.014	0.40	0.24	0.13
Ar	104 ± 10	0.134 ± 0.02	0.23	0.19	0.086
N ₂	55.2 ± 4.4	0.073 ± 0.015	0.22		

^aP. Violino (private communication).

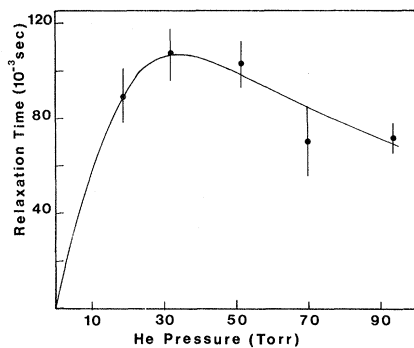


FIG. 5. Hyperfine relaxation time τ_0 of Cs vs He pressure. The points are experimental results and the continuous line is the theoretical fit.

The results obtained from the solution of this equation must be compared with the experimental measurements of τ_0 . A best fit for the parameters D_0 and $\sigma_{\langle \vec{S} \cdot \vec{I} \rangle}$ is not a trivial problem because of the complicated structure of Eqs. (3) and (4)—see also (A1) and (A2) in the Appendix. However we have developed an efficient computer program that searches the best parameter values with an iterative method by linear approximation of the function (3). With reasonable starting values for the parameters the method is rapidly convergent and typically only four or five steps are usually necessary. The best values for D_0 and $\sigma_{\langle \vec{S} \cdot \vec{I} \rangle}$ for each cesium-buffer-gas pair are listed in Table I. The theoretical curves obtained with these values are compared with experimental data in Figs. 5–8.

IV. ERROR ANALYSIS

We have considered the following causes as possible sources of error:

a. Systematic errors caused by an incorrect computation of cesium density. These errors are important in the measurement of σ_{ex} while they are practically negligible for τ_0 .

b. Influence of an external magnetic field on the value of τ_0 . No measurable effect of the magnetic field H_0 has been observed; it is worth noting that a dependence of Zeeman relaxation on H_0 has been observed⁹ in the case of Kr as a buffer gas while τ_0 in the same cells and under similar conditions does not change with H_0 .

c. Possible lowering of pumping efficiency at high buffer pressures caused by collisional broadening of the optical lines. The available data¹⁰ on the pressure broadening of Cs resonance lines are not in agreement with each other and one cannot therefore exclude such an effect. However in our measurements of Zeeman relaxation we have observed, for each buffer gas, a decrease in the pumping rate starting from pressures 1.5 or 2 times greater than

the maximum value used for hyperfine measurements.

d. Detection-beam problems. The intensity of the detection light beam is so small that it cannot cause any appreciable change in the observed relaxation time. The different geometry of a narrow detection beam is taken into account by suitable formulas as explained in the Appendix. This method of analysis has been checked in the following way: The results for D_0 and $\sigma_{\langle \vec{S} \cdot \vec{I} \rangle}$ obtained by Franzen's method in the case Cs-Ne have been used to compute the relaxation time one should observe in given conditions with a narrow detection beam. The agreement between this computation and the time experimentally measured is always within 2–3%. Such a small deviation shows that the method is reliable and that the errors for D_0 and $\sigma_{\langle \vec{S} \cdot \vec{I} \rangle}$ quoted in Table I are perhaps overestimated.

e. Spatial distribution of the initial population difference. We have computed the spatial distribution of the observable $\langle \vec{S} \cdot \vec{I} \rangle$ at the beginning of the relaxation transient with the method described in Ref. 8. We assume that the effect of the pumping light is equivalent to a distribution of sources that, at every point of the cell, create a population difference proportional to the pumping light intensity at that point. This source distribution is assumed to be uniform within the cylinder defined by the pumping light beam and zero outside. In the temperature range of the measurements the cell is optically thin; the approximation of small optical thickness (i. e., constant pumping light intensity) becomes certainly good when the results are extrapolated to zero cesium density. So we think that the assumption of a uniform distribution of pumping sources cannot cause any significant error in the quoted values of

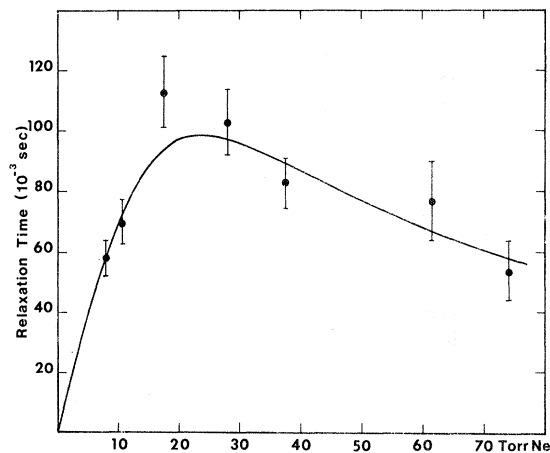


FIG. 6. Hyperfine relaxation time τ_0 of Cs vs Ne pressure. The points are experimental results and the continuous line is the theoretical fit.

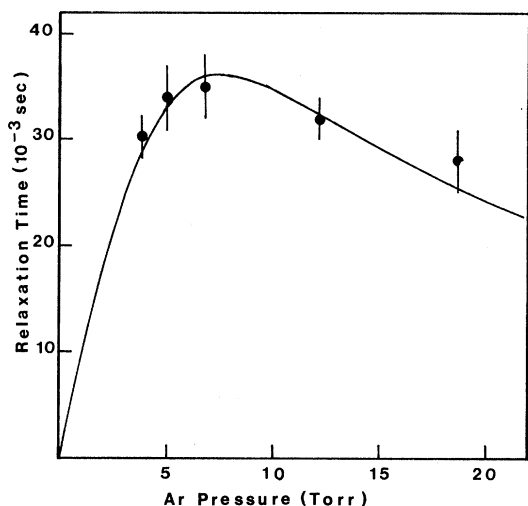


FIG. 7. Hyperfine relaxation time τ_0 of Cs vs Ar pressure. The points are experimental results and the continuous line is the theoretical fit.

D_0 and $\sigma_{\langle \vec{s} \cdot \vec{i} \rangle}$. The parameter, to which these values are more sensitive, is the pumping beam radius R_p . As the collimation of the pumping light cannot be perfect and its radial distribution is not exactly a step function, the dependence of D_0 and $\sigma_{\langle \vec{s} \cdot \vec{i} \rangle}$ on the R_p value must be tested. We have performed this test by changing the input parameter R_p in the computer program; the errors quoted in Table I take into account the effect of possible R_p fluctuations.

V. DISCUSSION

Let us compare the results and the meaning of the hyperfine relaxation study with the results of Zeeman relaxation in cesium. It is experimentally observed that the relaxation transient for $\langle \vec{S} \cdot \vec{I} \rangle$ is a single-exponential function and, therefore, the relaxation time is unequivocally defined. For $\langle S_z \rangle$ the decay has a more complicated shape^{11,12} and in most cases one can describe it, within experimental errors, as the sum of two exponential functions of different amplitudes: One can therefore define two phenomenological time constants τ_1 and τ_2 . This is not a surprising result insofar as because of the nuclear spin effect, the number of sublevels involved in Zeeman relaxation is $2(2I+1)$ while it is practically only 2 for hyperfine relaxation.

The presence of two (or more) time constants makes the diffusion problem rather complicated: A satisfactory method for dealing with the two time constants τ_1 and τ_2 together with the solution of a diffusion equation is not available.

In principle one might use the method proposed in Ref. 13 but it is difficult to decide whether the two

observed time constants τ_1 and τ_2 are caused by different diffusion modes or by different observables; usually an intermediate situation is encountered. So the fitting problem is not well defined and the results become unreliable.

Experimental works about electron relaxation of cesium in the presence of Ne, Ar, and N_2 have been published by Franz and Lüscher¹⁴ and for Ne, He, and Ar by Legowski.¹⁵ Franz and Lüscher have considered only the longest time constant¹⁶ and compared it with the decay time of the first diffusion mode. Legowski on the contrary has ascribed the two time constants to the first and the second diffusion mode and he has assumed that the Cs-buffer interaction is described by a single relaxation time. Both of these methods are not completely satisfactory on the basis of an oversimplified data interpretation and of a reduction of the number of parameters and they are acceptable only as a first approximation. The values of D_0 from Refs. 14 and 15 are listed in Table I for comparison purposes. They are larger than both the results of present paper and the theoretical values. A tentative explanation is the following: The measurements have been performed at a temperature (44 °C, Ref. 14) where diffusion modes higher than the first are important. The coefficient of D_0 in the expression of the time constants $\tau_{i\nu}$ of the higher modes increases rapidly with mode number [see Appendix, Eq. (A1)], so that an artificially large D_0 value is necessary to compensate for an imposed low value for the geometrical factor when only the first mode is used. With Legowski's method the values of D_0 are obviously smaller, but here the cesium-buffer-gas interaction is described by a single time constant. In

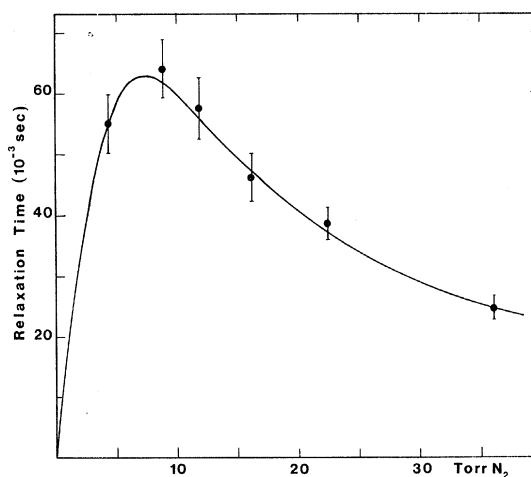


FIG. 8. Hyperfine relaxation time τ_0 of Cs vs N_2 pressure. The points are experimental results and the continuous line is the theoretical fit.

conclusion it seems reasonable that the D_0 value obtained from hyperfine relaxation measurements is more reliable than that obtained from Zeeman relaxation studies, because the situation is simpler at least from a phenomenological point of view. A further interest in the measurements of D_0 and $\sigma_{\vec{s}, \vec{i}}$ is the hope that an accurate knowledge of these parameters together with $\langle S_x \rangle$ relaxation data may be useful in the understanding of the relaxation mechanism.

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APPENDIX

In this appendix we give a few detailed formulas which are useful in the computation of the relaxation times. We shall use the following notations: R and L are the radius and length of the cell, R_p and R_d are the radius of the pumping and detection light beams (they are supposed $\leq R$), $J_n(x)$ is the Bessel function of order n , μ_i is the i th zero of $J_0(x)$, p is the buffer-gas pressure, p_0 is the reference pressure (760 Torr), N_L is the Loschmidt number, and \bar{v}_{rel} is the mean relative cesium-buffer velocity.

The time constants τ_{iv} of the different diffusion modes for a cylindrical shape are defined by^{1,8}

$$\frac{1}{\tau_{iv}} = \left(\frac{\pi^2(2\nu+1)^2}{L^2} + \frac{\mu_i^2}{R^2} \right) D_0 \frac{p_0}{p} + \bar{v}_{rel} \sigma_{\vec{s}, \vec{i}} \frac{p}{p_0} N_L. \quad (A1)$$

The weights of the different modes are given by

$$B_{iv} = \tau_{iv} \frac{J_1(\mu_i R_p/R) J_1(\mu_i R_d/R)}{[\pi \mu_i (2\nu+1) J_1(\mu_i)]^2}. \quad (A2)$$

Equations (A1) and (A2) are obtained by solving the diffusion equation^{4,5}

$$\frac{\partial}{\partial t} \langle \vec{S} \cdot \vec{I} \rangle = D \nabla^2 \langle \vec{S} \cdot \vec{I} \rangle - K \langle \vec{S} \cdot \vec{I} \rangle,$$

with the boundary condition that $\langle \vec{S} \cdot \vec{I} \rangle$ is zero on the cell walls. It is also assumed that the spatial distribution of $\langle \vec{S} \cdot \vec{I} \rangle$ at the instant $t=0$, the beginning of the relaxation transient, is the one obtained with constant pumping rate within the cylinder, defined by the pumping light beam, and zero outside.⁸ This assumption is reasonable in conditions of low cesium density as it is always the case.¹² Equations (A1) and (A2) show that a different pressure value modifies both the time constants τ_{iv} and the coefficients B_{iv} . This is one of the reasons that make the fitting problem complicated so that a computer is required: It is not possible to decide whether a single diffusion mode is predominant for all pressure values.

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