

A MEASUREMENT OF THE SPIN-ROTATION COUPLING IN NaXe MOLECULES

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The mean spin-rotation coupling γN for NaXe molecules is measured and found to be in reasonable agreement with a recent theoretical expression for the coefficient γ .

It was first argued by Bouchiat et al. [1] that a dominant mechanism in the spin relaxation of alkali atoms in the presence of heavy rare gases Ar, Kr and Xe is the formation of van der Waals molecules with lifetimes on the order of 10^{-7} s. This has also been seen [2,3] to dominate the relaxation of nuclear spin polarized Xe in alkali vapor. The collisions responsible for the formation of these molecules require a third body. This can be either another noble gas atom or an otherwise non-reactive atom or molecule such as N_2 , which is also used for more efficient optical pumping. A van der Waals molecule will persist until another collision overcomes the small binding. Thus the mean lifetime τ of the molecule is proportional to the inverse of the third body pressure P . During this lifetime the electronic spin S of the alkali is coupled to the nuclear spin K of the noble gas and to the rotation N of the molecule. Present experimental data on both types of relaxation mentioned above can be explained by the following hamiltonian [3]

$$H = AI \cdot S + \gamma N \cdot S + \alpha K \cdot S + g_s \mu_B S \cdot H, \quad (1)$$

where the first term is the alkali hyperfine interaction and the final term is the Zeeman interaction. The term $\alpha K \cdot S$ can cause spin transfer between S and K . But $\gamma N \cdot S$ dominates the relaxation of S - i.e. most of the spin S is transferred to N rather than K .

Recent interest has been in understanding the spin-rotation coupling, and, in particular, the coefficient γ . An early attempt to calculate this was by Herman [4], whose expressions for γ are proportional to the fine structure splitting of the alkali atom.

Based on this theory one would expect Na to be much more effective than the heavier alkalis in spin polarizing Xe, but Bhaskar et al. were unable to spin polarize Xe with Na [5]. Although they did not directly measure the spin-rotation coupling, their experiment suggested a much stronger coupling than expected. Other experiments [6] with KXe, RbXe and CsXe have also failed to see the substantial variation in γ that this theory predicts. Wu et al. [7] have derived an expression for γ which seems to agree with the experimental estimates for various combinations of K, Rb and Cs with Ar, Kr and Xe. In this experiment we infer γ for the NaXe system and compare this with the theoretical formula in ref. [7].

One can infer γ by observing either the fast alkali relaxation [8] or the slower noble gas nuclear spin relaxation [3,6]. In both cases what is actually measured is " γN ", an appropriate average of the coupling over vibrational and rotational states of the molecule. In this work we measure γN by observing the spin relaxation of Na.

The apparatus is shown schematically in fig. 1. The sealed cells contained a small amount of Na metal, 3.1 Torr natural Xe and various amounts of N_2 . We used both aluminosilicate (Corning 1720) and Pyrex cells. Although the former glass is preferred for its resistance to the alkalis, we found that Pyrex cells could survive for an adequate period (more than 2 hours) under the conditions of this experiment. The cells were heated to between 180° and 200° C, at which temperature the sodium density, $[Na]$, is between 10^{12} and 3×10^{12} cm^{-3} . In this range the

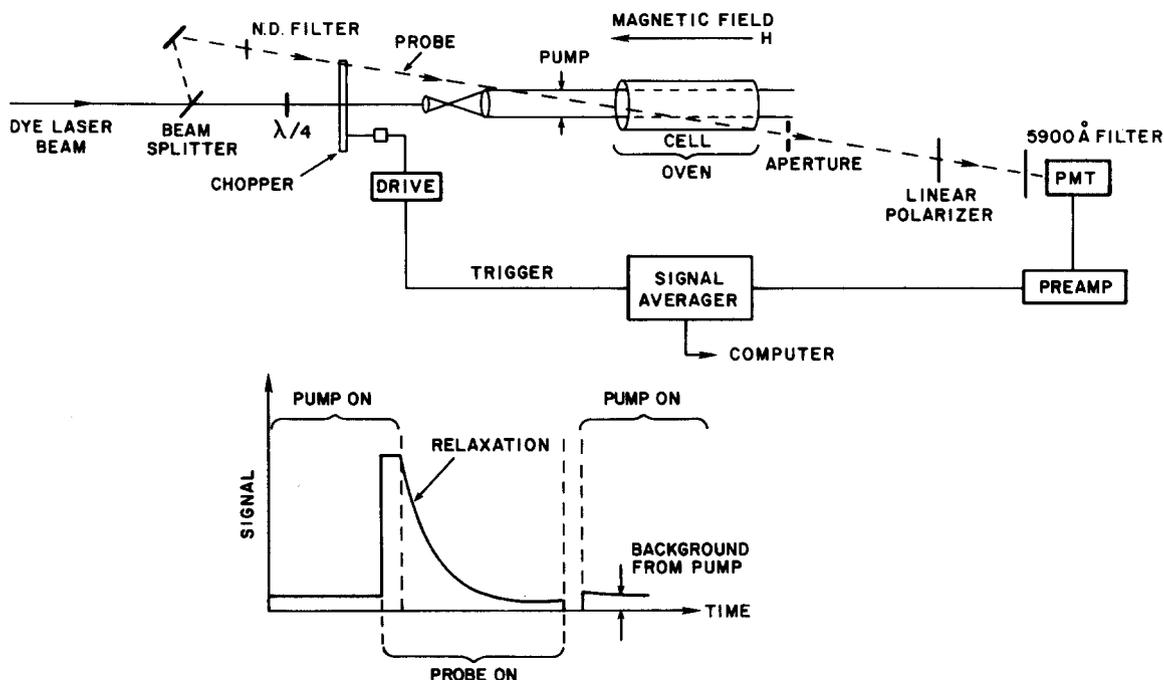


Fig. 1. A schematic of the apparatus and of the pump/probe cycle.

relaxation of S should be nearly independent of sodium density [5,9]. Thus only moderate temperature stability ($\lesssim 3^\circ\text{C}$ drift per hour) was adequate for our measurements. Both optical pumping and probing were done with a cw tunable dye laser (Spectra Physics model 365) using Rhodamine 6G dye. The dye was pumped by a 5 W argon ion laser (Spectra Physics model 165-03). Beam splitters were used to separate and attenuate the probe beam. The chopping cycle (fig. 1) was simple: both beams were on for half of each cycle with their "on" portions overlapping slightly at the beginning of the probe phase. The dye laser has a bandwidth of ~ 5 GHz, or about 100 times the Doppler broadening. Thus it was not possible to observe the relaxation by monitoring the absorption of circularly polarized light (circular dichroism technique) as in ref. [5] — there was simply too much background of off-resonant light. We observed spin relaxation with crossed linear polarizers, which attenuated this background.

There are a number of sources where the propagation of light in polarized vapors is discussed [10]. The relevant qualitative aspects are as follows. Due to preferential absorption of one circular polarization

over the other, linearly polarized incident light on resonance becomes elliptically polarized. While off-resonant light is not attenuated by the vapor, the two circular components σ_+ and σ_- will propagate with different phase velocities. This causes linearly polarized light to be rotated as it passes through the vapor. The angle θ of rotation of the plane of polarization is proportional to $\langle S_z \rangle$ to lowest order. Given the broad profile of the laser used here, it is this paramagnetic Faraday effect which is most important in our experiment.

For exactly perpendicular polarizers both resonant and off-resonant light give signals proportional to $\langle S_z \rangle^2$ in the limit of small θ , while the background of far-off-resonant light is maximally attenuated. But the polarizers are not perfect and it is hard to define perpendicular to better than a few tenths of a degree. This problem is compounded by Faraday rotations of as much as 8° for the magnetic fields used. For example, if the polarizer setting deviates by an angle δ from perpendicular, then the signal will be proportional to $\sin^2(\theta + \delta) \approx \theta^2 + 2\delta\theta + \delta^2$. This introduces a term linear in $\langle S_z \rangle$, which for sufficiently small $\langle S_z \rangle$ (long times), will dominate the quadratic term.

Roughly speaking frequencies above and below resonance are rotated in opposite directions. Thus for the laser profile centered near resonance, the term linear in $\langle S_z \rangle$ is very small. Still, in fitting the decay curves it was found that the rates were extremely sensitive to δ . Because of this difficulty, we wished to maximize the linear term while suppressing higher order terms. We accomplished this by tuning the laser to one side of resonance; then we took the difference of two signals with the polarizer displaced $5-10^\circ$ on each side of perpendicular. The result was almost purely linear in $\langle S_z \rangle$, and the rates determined in this way were insensitive to the choice of the polarizer settings.

Each transient signal was detected with a photomultiplier tube (RCA C7151W), amplified and averaged over 1024 periods with a Nicolet 2100 signal averager. The difference of the two signals above was recorded by an IBM PC/XT. Each of these differences was fit on the PC to a single exponential to determine the rate of decay.

Increasing magnetic fields H slow down the Xe induced relaxation rate of $\langle S \rangle$ by decoupling N and S [8]. For example, in high fields the Zeeman interaction dominates the spin-rotation interaction and S remains locked about H during the lifetimes of the molecule. The observed molecular induced rate as a function of H closely approximates a lorentzian with half width ΔH . We fit these slowing down curves to a function of the form:

$$\theta^{-1} = \theta_w^{-1} + \frac{\theta_0^{-1}}{1 + (H/\Delta H)^2}, \quad (2)$$

where θ_w^{-1} represents the relaxation due to binary collisions and diffusion to the wall. θ_0^{-1} is independent of field H for the conditions of interest here [5]. An example for one of our cells is shown in fig. 2. The width ΔH gives information about the lifetime τ and about the spin-rotation coupling. One expects that the width ΔH should increase with third body pressure P since the molecular lifetime decreases. According to the theory by Happer et al. [11] the width ΔH for high P is given by

$$\Delta H = (2I + 1)\hbar/\mu_B g_S \tau = \beta P, \quad (3)$$

where

$$\beta = (2I + 1)\hbar/\mu_B g_S \tau P \quad (4)$$

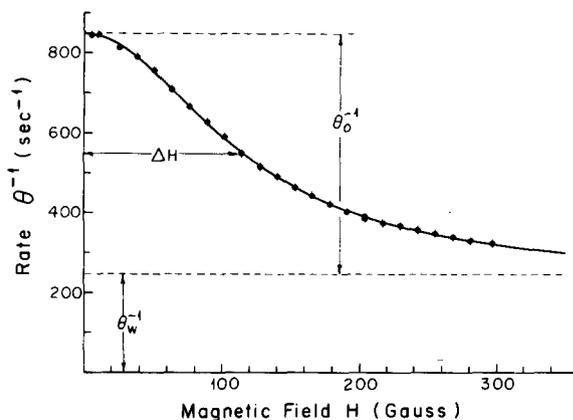


Fig. 2. The solum electronic spin relaxation rate versus magnetic field for one of our cells. The solid line is the fit.

is a constant. There is a nonzero limiting value ΔH_0 for small P (or equivalently long τ). Interpolation between the extremes is done through the formula

$$(\Delta H)^2 = (\Delta H_0)^2 + \beta^2 P^2. \quad (5)$$

Fig. 3 shows our data. Two different buffer gases will in general have different efficiencies (i.e. different β). For example in the RbXe system it was found [12] that Xe is roughly 3 times more effective than N_2 , i.e. $\beta_{Xe} \approx 3\beta_{N_2}$. We assume that this relative efficiency for the NaXe system is in the range 1-4, which

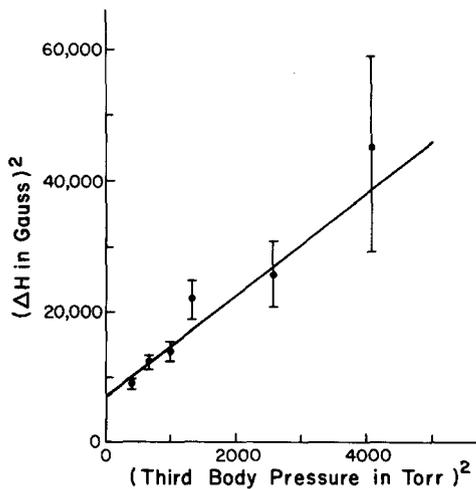


Fig. 3. $(\Delta H)^2$ versus P^2 for various pressures P . We have shown the data for $\beta_{Xe} = 3\beta_{N_2}$ (i.e. the equivalent N_2 pressure = $P_{N_2} + 3P_{Xe}$).

corresponds to an uncertainty of about 10% in the parameters ΔH_0 and β . We find

$$\Delta H_0 = 83 \pm 10 \text{ G}, \quad \beta = 2.8 \pm 0.3 \text{ G Torr}^{-1}.$$

Relation (4) above gives us the lifetime—pressure product for the molecules (in the presence of N_2) $\tau P = (8.1 \pm 0.8) \times 10^{-8} \text{ s Torr}$. It was found earlier [3,6] that such products for KXe, RbXe and CsXe molecules are all about $1.4 \times 10^{-7} \text{ s Torr}$.

In the limit of small P the width ΔH_0 is related to γN by [11]

$$\Delta H_0 = \gamma N / g_s \mu_B h_0, \quad (6)$$

where h_0 is the half width of the alkali spin transfer coefficient as a function of the dimensionless field parameter h

$$-q(F, F) = \frac{h^2 + 1 + (4/3x^2)K(K+1)}{4h^2} + \frac{(4/3x^2)K(K+1) - h^2 + 1}{16h^3} (h^2 - 1) \ln \left(\frac{h+1}{h-1} \right)^2.$$

Actually h_0 depends only weakly on K and $x = \gamma N / \alpha$, the Breit–Rabi parameter. Given the isotopic composition of natural Xe and assuming values of x somewhat larger than those found for K, Rb and Cs (as we expect), we estimate $h_0 = 1.35 \pm 0.05$. With this estimate

$$\gamma N / h = 172 \pm 22 \text{ MHz}.$$

By the method of ref. [1] we obtain $N_{\text{rms}} = 42$. From this we get ($\gamma N = \gamma_{\text{exp}} N_{\text{rms}}$)

$$\gamma_{\text{exp}} / h = 4.1 \pm 0.5 \text{ MHz}.$$

We see in fig. 4a that our value of γN is nearly the same as was found for K, Rb and Cs [6]. Since N is larger for heavier alkalis, the variation in γ_{exp} is more pronounced (fig. 4b). But it is clear that these data do not reflect the great variation in the fine structure splittings that the theory of Herman would imply [4] — in fact γ_{exp} decreases with atomic number. Although γ_{exp} for NaXe is greater than the value predicted by the recent theory [7]

$$\bar{\gamma}_{\text{th}} / h \approx 2 \text{ MHz},$$

one must bear in mind that γ is a very sensitive function of R , the radial separation of the molecule. The

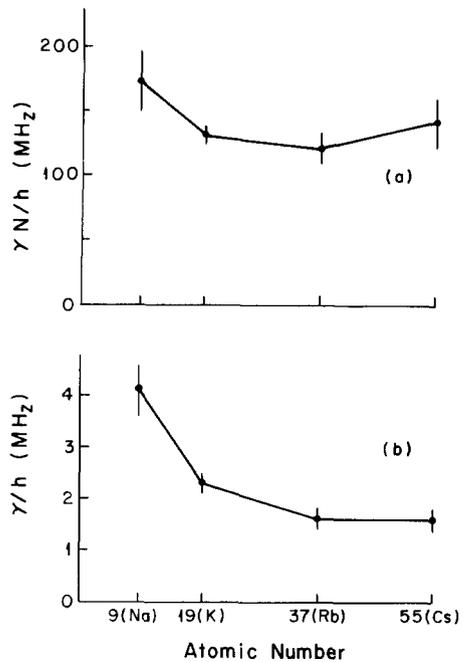


Fig. 4. Experimental γN and γ for the various alkalis in conjunction with Xe. The values of γN and γ for KXe, RbXe and CsXe are taken from refs. [6,7] with corrections due to binary collisions and the distribution of N .

value above is based on an equilibrium separation $R_0 = 5.03 \text{ \AA}$ [13], but a value just 5% lower than this almost doubles $\bar{\gamma}_{\text{th}}$. Also the “mean” $\bar{\gamma}_{\text{th}}$ is obtained from $\gamma(R)$ in a very crude way [7]. It is interesting to note that γ/h calculated at the equilibrium separation $R_0 = 5.03 \text{ \AA}$ (without averaging) is 4.0 MHz, according to this theory.

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