

WALL RELAXATION OF SPIN POLARIZED ^{129}Xe NUCLEI

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The wall relaxation of spin polarized ^{129}Xe nuclei is much longer in silicone coated pyrex cells than in uncoated cells. In contrast to uncoated glass cells where the wall relaxation times are unpredictable and usually only a few tens of seconds, the relaxation time of ^{129}Xe in silicone-coated cells is usually 20 minutes or longer.

We have found that silicone coatings on the interior surface of glass cells can substantially increase the wall relaxation times of ^{129}Xe gas in the cells. Recent experiments have shown that ^{129}Xe nuclei can be efficiently polarized by spin exchange with optically pumped alkali vapors [1–5]. The polarized ^{129}Xe nuclei relax due to spin interaction in loosely bound noble-gas–alkali-atom Van der Waals molecules. The molecular contribution to the relaxation can be suppressed by external magnetic fields of a few hundred Gauss. A much smaller contribution to the relaxation is made by binary collisions between the noble gas and alkali atoms [4]. Inhomogeneities in the external magnetic field can substantially shorten the relaxation time. Finally, the container walls dominate the relaxation rate in the absence of magnetic field inhomogeneities and at low alkali vapor density.

We have made extensive studies of ^{129}Xe mixed with ^{87}Rb vapor and various amounts of nitrogen or helium gas in spherical pyrex cells, 30 mm in diameter. We have fabricated nearly 150 cells. We find that the performance of uncoated pyrex cells is highly variable, with most wall relaxation times on the order of a few tens of seconds. The relaxation times sometimes become appreciably longer when the cell is heated to about 80°C and cooled to room temperature repeatedly. By baking newly manufactured cells at 80°C for a few weeks it is sometimes possible to lengthen the wall relaxation time to several hundred seconds. In one exceptional case an uncoated cell was observed to have a relaxation time of 1200 s. This

cell appears to have been accidentally contaminated with a few droplets of a transparent liquid, possibly silicone pump oil or kerosine from the calcium chips used to reduce the $^{87}\text{RbCl}$ during cell filling. We have not been able to determine the cause of this erratic cell behavior and we have been seriously hampered in our studies of gas phase relaxation by the low yield of useful cells with uncoated glass walls.

We find that pyrex cells which have had their interior walls treated with a commercial silicone coating agent (surfrasil)^{*1} are much superior to uncoated cells for studies of nuclear spin polarized ^{129}Xe . We always find ^{129}Xe wall relaxation times of 20 minutes or longer in these cells. The cell preparation is very simple. A mixture of 90% cyclohexane and 10% surfrasil is injected into a spherical pyrex cell, 30 mm in diameter, through a 3 cm stem. After shaking the solution in the cell, the excess is drained off and the cell is allowed to dry in air for several days. The cell is then sealed onto a glass manifold, baked at 150°C overnight under vacuum (2×10^{-6} Torr) and filled with xenon, a few droplets of distilled alkali metal and an appropriate amount of buffer gas (e.g. N_2 or He). In melting off the cell stems with a torch we limit the region of heated glass as much as possible to avoid damage to the coating.

The apparatus used to measure ^{129}Xe relaxation

*1 We use the commercial siliconizing agent *surfrasil* manufactured by the Pierce Chemical Company, Rockford, IL 61105, USA.

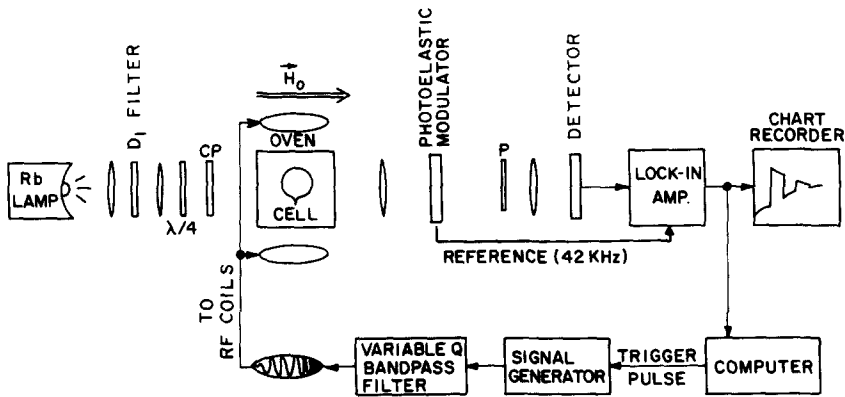


Fig. 1. A schematic of the experimental apparatus.

times is shown schematically in fig. 1. The sample is contained in a cylindrical glass oven, heated with flowing hot air. A Rb vapor resonance lamp is used to pump the ^{129}Xe - ^{87}Rb mixture in the cell with circularly polarized 7947 Å (D1) light. A representative pumping time is 15 minutes. During the probe phase of the experiment the circular polarizer, CP, is removed, and the small amount of elliptical polarization introduced in the unpolarized probe beam by polarized ^{87}Rb atoms is detected with a photoelastic modulator [6]. The signal is proportional to the number density of ^{129}Xe spin-polarized nuclei since the short lived (~ 1 ms) polarized ^{87}Rb atoms are produced by spin exchange with the xenon. Small amounts of stray elliptical polarization due to bire-

fringence of the lenses, oven windows and other optical components are eliminated with a rotatable plastic $\lambda/4$ plate. These stray components of polarization are quite temperature sensitive, so stabilization is important. Slow drifts in the system are often a problem for the very long ($\sim 1/2$ h) decay times which characterize some of these experiments. To eliminate errors due to the slow drift we perform adiabatic rapid passage [7], i.e. we periodically invert the ^{129}Xe nuclear polarization with a chirped audio-frequency magnetic field. In our experiments we operate at a static magnetic field of 5 Gauss, low enough to cause negligible slowing down of the relaxation. The chirped frequency is swept from 3 kHz to 7 kHz through the ^{129}Xe magnetic resonance frequency of 5 kHz. The chirping

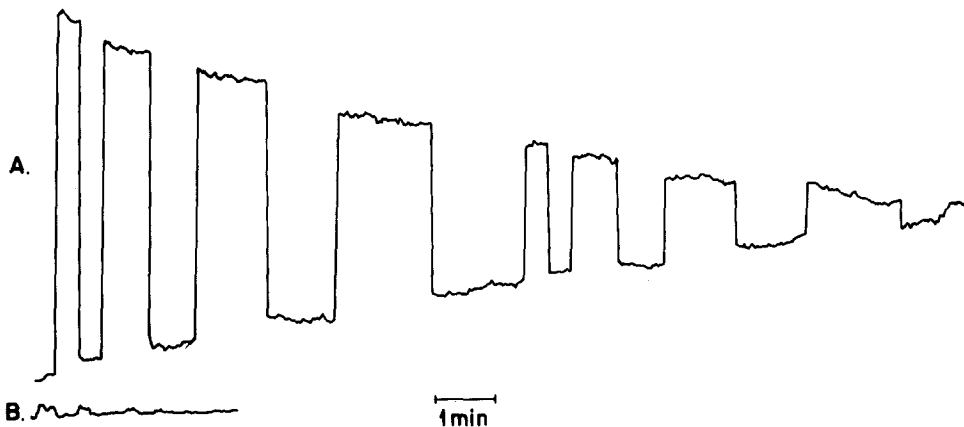


Fig. 2. ^{129}Xe decay curves. (A) Coated cell with 98.5 Torr He. (B) Uncoated cell with 91.5 Torr He.

rate is controlled by an Apple II plus computer which is also used for on-line data processing.

The Xe polarization at the time of an inversion is determined by subtracting the average signals during the five second intervals before and after the inversion. This procedure is equivalent to a filtering operation which eliminates noise frequencies with periods longer than 10 s. The signal amplitude is given by $A(t) = A(0)(1 - \epsilon)^n \exp(-t/\tau)$, where ϵ is the decay per swing (polarization inversion), n is the number of swings, and τ is the ^{129}Xe relaxation time. Variation of the interval between swings during the measurement permits the determination of both τ and ϵ from the decay curve.

Representative decay curves for ^{129}Xe are shown in fig. 2 for a coated and an uncoated cell.

The wall relaxation time in the uncoated cell is about 30 s, but it is about 2000 s in the coated cell. The fast wall relaxation reduces the number density of spin-polarized ^{129}Xe during the pumping cycle, thus the signal amplitude is much smaller. The advantages of a coated cell can be clearly recognized from fig. 2. To account for the effects of Rb-induced gas phase relaxation we measure the relaxation time in each cell at various temperatures and use the saturated vapor pressure formula [8] for Rb to extrapolate the relaxation rate to zero number density, as shown in fig. 3 for a coated cell and for an exceptionally good uncoated cell. The different slopes result from the difference in N_2 pressure [4]. We should note that at higher cell temperatures ($T \sim 80^\circ\text{C}$) coated cell walls seem to react with Rb vapor. We find that unless a large fraction of the coated cell wall is covered by Rb metal droplets the Rb vapor pressure within the cell is well below the value one would infer from the cell temperature.

A partial summary of our measurements is shown in fig. 4.

In conclusion we note that the fundamental reason for the substantial increase in the relaxation times of ^{129}Xe in coated cells is not yet understood, although several possibilities suggest themselves. The heat of adsorption of Xe atoms on the cell coating may be substantially less than on bare glass or on alkali metal. Thus, a xenon atom in a coated cell may be much less likely to be adsorbed on the wall and it will therefore suffer less wall relaxation no matter what the ultimate source of spin interactions on the wall should

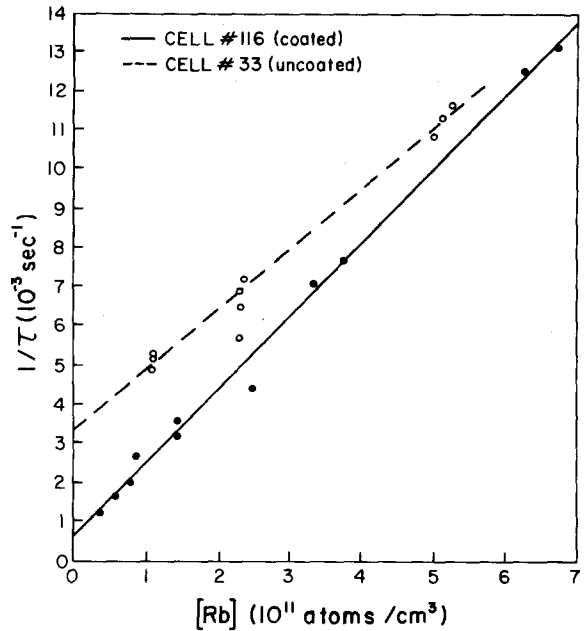


Fig. 3. ^{129}Xe decay rates as a function of Rb density. The nitrogen filling pressure is 21 Torr for the uncoated cell and 14.9 Torr for the coated cell.

be. Alternatively, much of the relaxation may be Korringa relaxation [9] of ^{129}Xe nuclei adsorbed on the exposed surface of the alkali metal within the cell. The cell coating may prevent the formation of a continuous film of alkali metal and force the alkali metal to condense into a relatively small number of droplets. If less alkali-metal surface area is available there will be less Korringa relaxation. It is also possible that the surface coating buries paramagnetic sites within and near the surface of the glass so the ^{129}Xe never gets near enough to the site to relax effectively. Large numbers of dangling bonds and

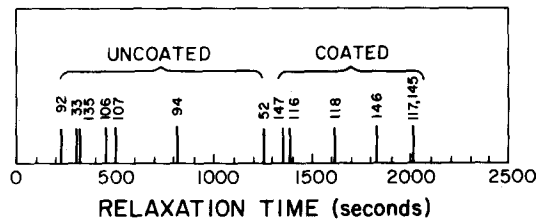


Fig. 4. Wall relaxation rates of ^{129}Xe for various coated and uncoated cells. The serial number of the cell is noted on the graph.

other paramagnetic sites are known to exist at silica surfaces under some conditions [10]. Preliminary experiments in our laboratory indicate that wall relaxation times of one hour or more can be obtained if the alkali metal is confined to a small area within the stem of the cell.

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