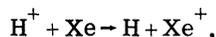


Table I. Energy dependence and  $E\theta$  values for the two sets of minima indicated in Fig. 1.

Energy (eV)	$\theta_A$ (deg)	A $10^{-2}\theta_A E$	$\theta_B$ (deg)	B $10^{-2}\theta_B E$
70	2.9	2.05	7.6	5.3
100	2.0	2.0	4.9	4.9
150	1.4	2.1	3.5	5.2
200	...	...	2.8	5.6

of our hypothesis. Also a comparison with the corresponding elastic-scattering cross section would be very interesting.

No comparable structure was found for the reaction



which is exothermic by only 0.16 eV if the elec-

tron is transferred to the first excited state of the  $\text{Xe}^+$  ion.

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### VERY LONG NUCLEAR SPIN RELAXATION TIMES IN GASEOUS $\text{He}^3$ BY SUPPRESSION OF $\text{He}^3$ -SURFACE INTERACTIONS\*

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Experiments are described that reveal the nature of the surface relaxation mechanisms between  $\text{He}^3$  atoms and their containing walls, and effective means for their suppression. The resultant very long  $\text{He}^3$  relaxation times ( $>2 \times 10^5$  sec at 300°K, and rising rapidly with temperature) measured in highly spin-polarized samples prepared by optical pumping offer new possibilities for high-density polarized  $\text{He}^3$  nuclear targets, and for experiments requiring near-perfect gyroscopes.

Interest in nuclear spin relaxation mechanisms in  $\text{He}^3$  gas has been stimulated in recent years by the development of optical pumping techniques for dynamically polarizing the  $\text{He}^3$  nuclei.<sup>1</sup> The intrinsic spin relaxation time resulting from nuclear dipole-dipole interactions in  $\text{He}^3$  gas is reliably estimated to be of order  $4 \times 10^7 \times T^{1/2} \rho^{-1}$  sec,<sup>2</sup> where  $T$  is the absolute gas temperature and  $\rho$  the density expressed in units equal to the sample pressure in Torr at room temperature. However, observed relaxation times have been orders of magnitude shorter than this because of interactions of  $\text{He}^3$  atoms with the containing walls.<sup>1,3,4</sup>

The experiments reported here were undertaken in an effort to elucidate the nature of the  $\text{He}^3$ -surface interaction and, if possible, to find means to suppress surface relaxation ef-

fects. As a result of these studies two distinctly different  $\text{He}^3$ -surface interactions have been identified, and it was found that at moderate temperatures surface relaxation of  $\text{He}^3$  gas can be suppressed to the extent desired by judicious selection of container materials. The resulting very long  $\text{He}^3$  nuclear spin relaxation times observed suggest new avenues for development of nuclear gyroscopes and dense polarized  $\text{He}^3$  targets for use in nuclear scattering experiments.

The experiments were performed on gaseous  $\text{He}^3$  samples contained in sealed spherical glass bulbs of approximately 90-cc volume. The glass sample containers were first cleaned chemically, then heated under vacuum and subjected to helium electrical discharge cleaning in a manner described by Colegrove, Schearer,

and Walters.<sup>1</sup> He<sup>3</sup> gas at the desired pressure was then admitted to each sample cell prior to sealing. Sample densities varied over a range corresponding to gas pressures of about 1 to 20 Torr at room temperature.

Nuclear spin relaxation times were measured by first inducing a large (~10%) nuclear polarization in the discharge-excited sample by the optical pumping method.<sup>1</sup> The discharge excitation was then removed and the time rate of decay of the polarization to its thermal equilibrium value in a field of 200 G was monitored by a conventional nuclear magnetic resonance technique.

Typical results of relaxation time measurements for He<sup>3</sup> samples in Pyrex and Suprasil quartz containers are shown in Fig. 1 as a function of temperature. Three curves for Pyrex containers are shown to indicate the variation from sample to sample in absolute magnitude of the measured relaxation times. Measurements on a given sample are reproducible. No systematic variation of relaxation time with density was observed; this is to be expected for surface-induced relaxation since the wall collision rate is proportional to density.<sup>5</sup>

The most interesting feature of the data shown in Fig. 1 is the characteristic relaxation time maximum occurring near 125°K. This behavior was observed for all samples contained in Pyrex and quartz, and it suggests that distinctly different surface relaxation mechanisms

are operative at lower and higher temperatures. A phenomenological model that describes the data satisfactorily can be developed in terms of a low-temperature mechanism that involves physical adsorption of He<sup>3</sup>, and a high-temperature mechanism involving permeation of He<sup>3</sup> into the bulk of the container material.<sup>6</sup>

In terms of the average sticking time  $t_s$ , the number  $n$  and relaxation time  $T_s$  of He<sup>3</sup> atoms stuck on the surface, and the total number  $N$  of He<sup>3</sup> atoms in the container, it can be shown that the surface-induced nuclear spin relaxation time  $T_1$  of the He<sup>3</sup> gas is given by

$$T_1 = (N/n)(T_s + t_s) \quad (1)$$

under the assumption that  $n \ll N$ .<sup>7</sup>

At low temperatures, where sticking by adsorption predominates,  $t_s \approx t_s^0 \exp(E/kT)$ , where  $E$  is the energy of adsorption (~0.01 eV)<sup>8</sup> and  $t_s^0$  a constant of order  $10^{-13}$  sec.<sup>9</sup> The number of atoms adsorbed is just the product of  $t_s$  and rate at which atoms strike the wall; i.e.,  $n = (N\bar{v}A/4V)t_s$ , where  $V$  and  $A$  are, respectively, the volume and surface area of the container, and  $\bar{v}$  is the mean thermal velocity of the He<sup>3</sup> atoms. For  $T_s \gg t_s$  (a reasonable assumption for  $E < kT$ ), Eq. (1) gives for the relaxation time  $T_{1L}$  in the low-temperature limit

$$T_{1L} \approx 4(VT_s/A\bar{v}t_s^0) \exp(-E/kT). \quad (2)$$

At higher temperatures the He<sup>3</sup> atoms are able to diffuse into the container walls. Dissolved He<sup>3</sup> atoms within a diffusion jump distance  $\langle \Delta x \rangle$  of the surface are continually exchanging places with He<sup>3</sup> atoms impinging on the surface from the gas. The characteristic exchange time (i.e., the sticking time in the assumed model) is  $t_s \approx \alpha\tau$ , where  $\tau$  is the mean time between jumps for a dissolved He<sup>3</sup> atom, and  $\alpha \approx 6$  is a geometrical factor required because dissolved atoms may jump in directions other than toward the surface.  $\tau$  and  $\langle \Delta x \rangle$  are related through the diffusion coefficient  $D$  of helium in the glass of interest,  $D = \langle \Delta x^2 \rangle / 6\tau$ . For diffusion with activation energy  $Q$ ,  $\tau = \tau_0 e^{Q/kT}$ , where  $\tau_0$  is a constant for a given glass.<sup>10</sup> The number of dissolved atoms within a diffusion jump distance of the surface (i.e., those undergoing exchange) is  $n = SA\langle \Delta x \rangle NkT/V$ , where  $S$  is the solubility of helium in the glass of interest. Equation (1) gives for the He<sup>3</sup> gas

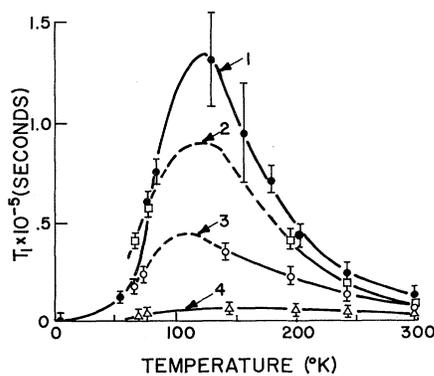


FIG. 1. Nuclear spin relaxation time versus temperature for He<sup>3</sup> samples in 90-cc spherical containers of Pyrex and quartz. Curves 1-3 represent the measurements on three samples in different Pyrex containers. Curve 1 is for sample pressure at room temperature of 10 Torr; Curves 2 and 3, for two different samples both at 20 Torr. Curve 4 represents the data taken on a single 10-Torr sample in a Suprasil quartz container.

relaxation time  $T_{1H}$ , at high temperatures,

$$T_{1H} = (V/A\langle\Delta x\rangle SkT)[T_s + \tau_0 \exp(Q/kT)]. \quad (3)$$

For Pyrex,  $S \approx 0.006$  (cc STP)/cc atm,  $Q = 0.28$  eV, and  $\tau_0$  is approximately  $10^{-13}$  sec assuming a jump distance of about 3 Å. The corresponding values for quartz are 0.01 (cc STP)/cc atm, 0.21 eV, and  $10^{-11}$  sec.<sup>10,11</sup> The magnitude and temperature variation of  $T_s$  are unknown, but  $T_s$  can be expected to decrease monotonically with temperature.

The predicted temperature dependence of surface-induced spin relaxation is then approximately given by

$$T_1^{-1} = T_{1L}^{-1} + T_{1H}^{-1}. \quad (4)$$

Equation (4) exhibits a relaxation-time maximum at intermediate temperatures, in agreement with the experimental data for Pyrex and quartz surfaces.

If the phenomenological model presented above is valid, one should expect an entirely different behavior for  $T_1$  and very long  $\text{He}^3$  spin relaxation times at the higher temperatures if an impermeable container material is used instead of Pyrex or quartz. This prediction of the model was tested by measuring the relaxation times of  $\text{He}^3$  contained in Corning 1720 and 1723 aluminosilicate glasses, both of which are known to be highly impermeable to helium.<sup>11</sup> The results are shown in Fig. 2 for representative samples; a typical set of data for a Pyrex container is shown for comparison. As expected, there is no evidence of permeation-dominated relaxation, so that  $T_1$  increases very rapidly with temperature in agreement with Eq. (2). Variations in the absolute magnitude of  $T_1$  from sample to sample presumably result from varying density of paramagnetic centers contributing to wall relaxation (e.g., through  $T_s$ ). However, more than half the samples exhibited relaxation times longer than  $10^5$  sec at room temperature; for these samples we establish only a lower limit for the surface-controlled relaxation times because magnetic-gradient relaxation<sup>3</sup> determines the measured  $T_1$  in our apparatus for  $T_1 \geq (1 \text{ to } 2) \times 10^5$  sec. There is every indication that much longer relaxation times can be realized at higher temperatures in a magnetic environment where gradients are minimized.

These measurements suggest that surface-induced nuclear spin relaxation in gaseous  $\text{He}^3$

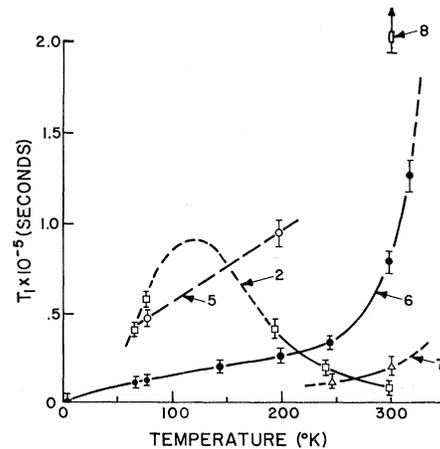


FIG. 2.  $\text{He}^3$  nuclear spin relaxation times for  $\text{He}^3$  samples in 90-cc spherical aluminosilicate glass containers. Curves 6 and 7 represent measurements on two samples in different containers made of Corning 1723 glass, with sample pressures at room temperature of 12 and 15 Torr, respectively. Curve 5 represents data taken on a single 10-Torr sample contained in Corning 1720 glass. Curve 2 is reproduced from Fig. 1 to facilitate comparison of Pyrex and aluminosilicate surface effects. The longest room-temperature relaxation time measured was on a 10-Torr sample in a Corning 1723 container, indicated by the point labeled 8; the observed  $T_1$  is determined by magnetic field gradients, and hence gives only a lower limit for the surface-controlled relaxation time.

can be suppressed to the extent desired by controlling the sample temperature and using impermeable container materials. It is not unreasonable to expect that intrinsic  $\text{He}^3$  spin relaxation times of the order of weeks to years (depending on sample density) can be achieved. Furthermore, such samples can be highly spin polarized by optical pumping<sup>1</sup> to yield appreciable macroscopic nuclear magnetization even in low-density  $\text{He}^3$  gas. These properties should be useful in applications requiring very low-friction gyroscopes (e.g., tests of Einstein's theory of relativity,<sup>12</sup> search for a  $\text{He}^3$  nuclear electric dipole moment<sup>13</sup>).

Suppression of surface relaxation effects also opens new possibilities for producing sizable nuclear polarization in dense  $\text{He}^3$  gas. A number of years ago Bouchiat, Carver, and Varnum attempted to polarize  $\text{He}^3$  gas at  $50^\circ\text{C}$  and 3-atm pressure by spin exchange with optically pumped rubidium vapor.<sup>14</sup> Though an effect was noted, the method failed to yield useful values of polarization because of the short-cir-

cutting effect of  $\text{He}^3$  spin relaxation at the container walls. For these experiments  $T_1$  was about  $10^3$  sec. We estimate that with the much slower wall relaxation to be expected from impermeable container materials, a polarization of the order of 10% might now be possible by this method. Of course such an estimate ignores the (so far unknown) effect of rubidium on the surface relaxation behavior. We are currently investigating this question and plan to repeat the experiment of Bouchiat, Carver, and Varnum.

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†National Aeronautics and Space Administration Fellow in physics.

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## TEMPERATURE DEPENDENCE OF POSITRON MEAN LIVES IN METALS

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The dependence of positron lifetimes on sample temperature has been studied in several metals over a range from room temperature to the melting point or to 400°C. Marked effects, amounting to as much as a 30% increase in lifetime, are noted in most of the low-melting-point metals. Lattice vacancies are considered to be the likely cause of these effects.

The lifetime of positrons in certain metals exhibits a marked dependence on sample temperature in the range from room temperature to 400°C. This result is closely related to the previously reported temperature dependence of the angular correlation of annihilation radiation.<sup>1</sup> We attribute the effect to lattice defects induced by temperature, and hence, it is also related to the recently observed dependence of both lifetime<sup>2</sup> and angular correlation<sup>3</sup> on plastic deformation.

The measurements reported here were obtained on a fast-slow coincidence system employing an overlap time-to-amplitude converter, pulse-height compensation, and pile-up rejection. With energy selection appropriate for lifetime studies, the system yielded a resolution of 410 psec on a  $^{60}\text{Co}$  source, the approximately exponential slopes corresponding to mean lifetimes of 90 and 103 psec.

All samples studied were of at least 4 nines

purity and were annealed and etched before insertion in the Pyrex sample chamber which operated with a helium atmosphere. The chamber was heated externally and a cooling system ensured that the scintillators and photomultipliers were not allowed to introduce false temperature-dependent effects. The simplest proof of the effectiveness of these precautions is in the markedly different results observed with the various metals but we also measured the temperature in the detector enclosures and held the variation below 10°C.

We observed two components in the decay of each metal. This is invariably the case when using a sandwich arrangement with a separate source between two samples, but it has been clearly demonstrated that the low-intensity tail is due to surface effects.<sup>4</sup> Although the tail is not of primary interest, we must make a reasonably accurate measurement of its slope and intensity in order to evaluate the main slope.