

axis, mainly because of a small population of trapped particles. This also fits the observation of the high-temperature region near the magnetic axis in  $ST$ .<sup>4</sup>

The experiment with the dc octupole<sup>5</sup> showed that the diffusion is collisional at high densities ( $n > 10^9 \text{ cm}^{-3}$ ). The measured diffusion coefficient is approximately the classical value inside the separatrix. The value is much larger outside of the separatrix where  $\langle \Delta\psi^2 \rangle$  is expected to be large. More detailed measurements are underway to determine the scaling on the magnetic field.

In summary, a model is proposed to explain the experimental observations of the anomalous transport processes in Tokamaks and multipole devices. Since this type of transport is only weakly dependent on the magnetic field strength,

a large fusion device must be carefully engineered to minimize the magnetic perturbations.

\*Work supported by the U. S. Atomic Energy Commission under Contract No. AT(04-3)-167, Project Agreement No. 38.

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## Isotopic Impurity Tunneling in Solid $^4\text{He}$ <sup>†</sup>

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(Received 3 June 1971)

A study has been made of the motion of isotopic impurities in solid  $^4\text{He}$ . The tunneling frequency of  $^3\text{He}$  atoms with neighboring  $^4\text{He}$  atoms in the  $^4\text{He}$  lattice has been deduced from nuclear magnetic relaxation measurements. The volume variation of the  $^3\text{He}$ - $^4\text{He}$  tunneling rate is much greater than that of the  $^3\text{He}$ - $^3\text{He}$  tunneling rate (exchange).

Recent discussions by Andreev and Lifshitz,<sup>1</sup> Guyer and Zane,<sup>2</sup> and Balakrishnan and Lange<sup>3</sup> have suggested the existence of a set of many-body excitations, called mass fluctuation waves, associated with the motion of isotopic impurities in solid-helium crystals. Because of the large-amplitude zero-point motion in quantum crystals, there is an overlap of the atomic wave functions of atoms occupying adjacent lattice sites which permits a mutual tunneling of the atoms through the potential barrier of neighboring atoms. The pairs of atoms rotate about the hard-core sphere of repulsion. At sufficiently low temperatures and with sufficiently dilute isotopic impurities, the tunneling motion permits the isotopic impurity to be distributed over a large number of lattice sites, and, further, the impurity atom propagates through the lattice with a well-defined wave vector.<sup>1</sup> The striking decrease in the spin-lattice relaxation time  $T_1$  which has been measured at low temperatures in  $^3\text{He}$  crystals doped with  $^4\text{He}$  impurities<sup>4-6</sup> has been analyzed in terms of the thermal reservoir associated with the

mass-fluctuation waves of the  $^4\text{He}$  atoms.<sup>2,7,8</sup>

In the present work we have attempted to detect the characteristic tunneling motion of isotopic impurities by studying the nuclear magnetic relaxation of  $^3\text{He}$  atoms diluted in  $^4\text{He}$  crystals.

Let us construct a simple theory to account for the relaxation resulting from the tunneling motion. We begin by assuming that the  $^3\text{He}$  atoms travel through the lattice by a random walk to nearest-neighbor positions with a mean time  $\tau_{34}$  between steps.  $\tau_{34}^{-1}$  is the tunneling frequency between  $^3\text{He}$  atoms and  $^4\text{He}$  atoms in the  $^4\text{He}$  lattice. The correlation function for the fluctuations in the nuclear magnetic dipole field will thus be an exponential decay with a "correlation time"  $\tau_{34}$ . The problem of spin-lattice relaxation by translational diffusion in a solid has been studied by Torrey.<sup>9</sup> A simple modification of his expression for  $T_1$  yields

$$T_1^{-1} = (2M_2 x / \omega) \psi(k, y), \quad (1)$$

where  $M_2$  is the second moment of the resonance line for a rigid lattice with  $x = 1$ ,<sup>10</sup>  $x$  is the con-

centration of  $^3\text{He}$  atoms,  $\omega$  is the Larmor frequency, and

$$\psi(k, y) = yG(k, y) + 4yG(k, 2y),$$

where

$$G(k, y) = \int_0^\infty J_{3/2}^2(kx) \frac{1 - (\sin x)/x}{[1 - (\sin x)/x] + y^2} \frac{dx}{x}, \quad (2)$$

in which  $y = \frac{1}{2}\omega\tau_{34}$  and  $k$  is a parameter that, like  $M_2$ , depends upon the lattice structure. For the hcp lattice we have  $k = 0.743$  and for an hcp powder of crystallites,<sup>11</sup>  $M_2 = 22.60 \times 10^{10}/V^2 \text{ sec}^{-2}$ , where  $V$  is the molar volume in cubic centimeters. In the high-field limit,  $\omega\tau_{34} \gg 1$ , Eq. (2) may be easily evaluated and the expression for the spin-lattice relaxation takes on the form

$$T_1^{-1} = 1.70xM_2/\omega^2\tau_{34}. \quad (3)$$

Resing and Torrey<sup>12</sup> have also developed a theory for the transverse relaxation time  $T_2$  associated with translation diffusion in a solid. In the high-field limit their expression becomes

$$T_2^{-1} = 0.922xM_2\tau_{34}. \quad (4)$$

We may thus expect to detect the existence of the impurity tunneling motion through the measurement of spin-lattice relaxation times which vary as  $\omega^2$  and are independent of temperature.<sup>13</sup> Studies of the magnitude of  $T_1$  and  $T_2$  should then yield the parameter  $\tau_{34}$ . In the present work we report such measurements.

The relaxation rates  $T_1$  and  $T_2$  were measured using standard pulsed NMR techniques.<sup>14,15</sup> Solid  $^4\text{He}$  crystals with isotopic impurities of mole fraction  $x = 0.02$ ,  $0.01$ , and  $0.005$   $^3\text{He}$  in  $^4\text{He}$  were prepared at constant volume using the blocked-capillary technique. The pressure of the solid helium thus formed was measured using a capacitance strain gauge<sup>16</sup> and the molar volume was then determined from the known melting parameters of solid helium.<sup>17</sup> The solid was cooled to low temperatures using a dilution refrigerator.

Differences in the absolute susceptibility of the various specimens measured in the nuclear relaxation experiments indicated that the actual concentration of  $^3\text{He}$  in the solid within the NMR coil varied from that in the gas mixtures used; sometimes by as much as 50% for the mole fraction  $x = 0.01$  in the gas and by larger amounts for  $x = 0.05$ . Frequently the susceptibility of the solid specimen in the coil would be less than that of the liquid mixture before solidification, assuming a Curie-law behavior for both the

liquid and the solid. This variation in concentration was probably due to isotopic enrichment of the liquid phase in  $^3\text{He}$  as the solid was formed. A region of  $^3\text{He}$  enrichment could have existed in any portion of the sample chamber, either inside or outside of the NMR coil; and also there was possibly an inhomogeneity in the  $^3\text{He}$  distribution within the coil. For solid mixtures with  $x < 0.01$ , the specimen concentration within the coil was assigned on the basis of susceptibility measurements with an uncertainty of a factor of 2.

Between the melting temperature and about 0.7 K,  $T_1$  is strongly temperature dependent and passes through a sharp minimum at around 1 K. The details of this high-temperature behavior have been previously studied<sup>18,19</sup> and are attributed to the thermally activated motion of vacancies through the lattice. Below about 0.7 K,  $T_1$  becomes temperature independent and is not observed to change with decreasing temperature until the onset of isotopic phase separation, below about 0.2 K for our samples. Figure 1 shows the frequency dependence of  $T_1$  in the temperature-independent region for a number of different molar volumes formed from the gas mixture with mole fractions  $x = 0.01$  and  $0.02$ . The data

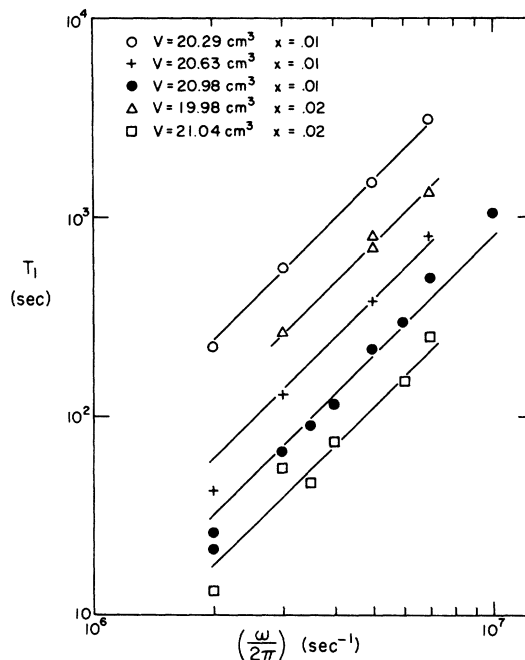


FIG. 1.  $T_1$  versus Larmor frequency at  $x = 0.01$  and  $0.02$ . Lines are typical of the  $\omega^2$  frequency dependence of  $T_1$  for  $0.02 > x > 0.005$ .  $T_1$  increases with decreasing molar volume and decreasing  $^3\text{He}$  concentration.

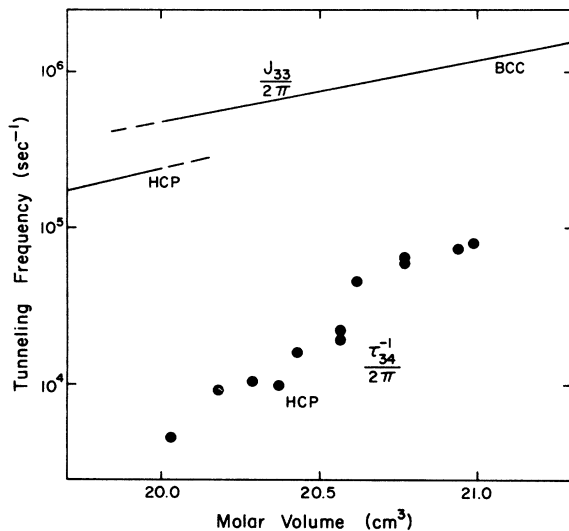


FIG. 2. Volume dependence of the tunneling frequencies. Points are obtained from  $T_1$  measurements at a Larmor frequency of 5 MHz with  $x = 0.01$ , using Eq. (3) in the text. Line is synthesis of available measurements of the  $^3\text{He}$ - $^3\text{He}$  tunneling frequency (exchange frequency). The impurity tunneling frequency,  $\tau_{34}^{-1}$ , has a much stronger volume dependence than the  $^3\text{He}$ - $^3\text{He}$  tunneling frequency,  $J_{33}$ :  $d \ln \tau_{34}^{-1} / d \ln V \approx 3 d \ln J_{33} / d \ln V$ .

in Fig. 1 show that  $T_1$  does indeed vary as  $\omega^2$ , as might be expected from Eq. (3).  $T_1$  in pure  $^3\text{He}$  and in the richer mixtures of  $^3\text{He}$  in  $^4\text{He}$  also displays a temperature-independent region which is due to the  $^3\text{He}$  tunneling with  $^3\text{He}$  neighbors (exchange). The frequency dependence of  $T_1$  for specimens rich in  $^3\text{He}$  is quite different from that observed here,<sup>19</sup> varying approximately as  $\exp(\omega^2/2x\omega_T^2)$ , where  $\omega_T$  is related to the exchange frequency  $J$  by  $\omega_T = 6.48J$ . The concentration dependence of  $T_1$  is qualitatively in agreement with Eq. (3).  $T_1$  increases as the concentration decreases. A precise determination of the concentration dependence is complicated by the scatter in the actual concentration of the specimens within the resonance coil.

Figure 2 shows the values of the parameter  $\tau_{34}^{-1}/2\pi$  at various molar volumes determined from  $T_1$  data measured at a Larmor frequency of 5 MHz with the mole fraction 0.01  $^3\text{He}$  in the gas used to form the solid. The points in Fig. 2 are calculated using Eq. (3). The scatter in the data is primarily due to the concentration variation rather than errors in molar volume (determined to better than 0.05  $\text{cm}^3$ ) or in the measurement of  $T_1$  (determined to better than 10%). The volume variation of the  $^3\text{He}$  exchange fre-

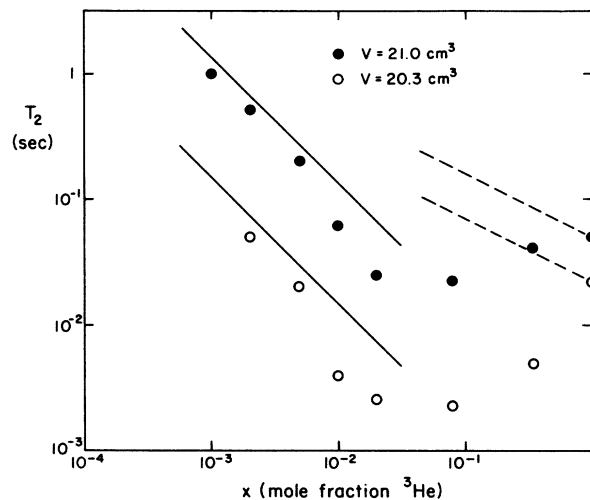


FIG. 3. The concentration dependence of  $T_2$ . The solid lines are calculated from Eq. (4) in text using values of  $\tau_{34}$  at 21 and 20.3  $\text{cm}^3$  obtained from Fig. 2. In the dilute limit,  $x < 10^{-2}$ ,  $T_2$  is consistent with the values expected for relaxation by means of  $^3\text{He}$ - $^4\text{He}$  tunneling. For  $x > 10^{-2}$ ,  $T_2$  is anomalously short. The dashed lines show the expected concentration dependence for relaxation by means of  $^3\text{He}$ - $^3\text{He}$  tunneling (Ref. 19). For  $x < 10^{-2}$  the concentration was determined from susceptibility measurements.

quency  $J/2\pi$  is also shown in Fig. 2 with solid lines obtained from the measurements<sup>20</sup> of  $(dP/dT)_V$  and a synthesis of nuclear relaxation data.<sup>21</sup> The most striking feature of the data in Fig. 2 is the very rapid increase in  $\tau_{34}^{-1}$  with volume compared with the change in  $J$  over the same volume range. There have been no theoretical predictions concerning the nature of the volume variation of  $\tau_{34}$ . The volume dependence of  $\tau_{34}$  determined from the concentrations of  $^3\text{He}$  other than  $x = 0.01$  is the same as that shown in Fig. 2.

The change in  $T_2$  with concentration is shown in Fig. 3 for the molar volumes 21 and 20.3  $\text{cm}^3$ . The data for mixtures richer than  $x = 0.02$  are taken from Miyoshi *et al.*<sup>19</sup> The decrease in  $T_2$  as pure  $^3\text{He}$  is diluted is surprising. The dashed lines indicate the  $x^{-1/2}$  increase in  $T_2$  expected<sup>19</sup> for the dilution effects upon the  $^3\text{He}$ - $^3\text{He}$  tunneling-relaxation mechanism. The solid lines are calculated from Eq. (4) using the values of  $\tau_{34}$  obtained from Fig. 2 to show the expected behavior of  $T_2$  in the very dilute limit in which the  $^3\text{He}$ - $^4\text{He}$  tunneling mechanism is responsible for the relaxation. For concentrations greater than  $x = 0.02$ , the actual behavior of  $T_2$  can be seen to be quite different from that predicted by these simple considerations. The

anomalously short values of  $T_2$  seem to imply that many of the  $^3\text{He}$  atoms are not free to experience the motional effects measured in  $T_1$  experiments on the same specimen. Eventually, as the specimen becomes dilute enough,  $T_2$  increases several orders of magnitude in a relatively narrow concentration range below  $x = 0.02$  to agree with the limiting value of  $T_2$  expected from the Torrey model.

Andreev and Lifshitz<sup>1</sup> suggested that when impurities exceed a critical concentration the interactions of impurities with each other through a crystalline strain field will impede the tunneling transition from site to site. Yamashita and Tsuneto<sup>8</sup> have estimated that the critical concentration above which the motion of dilute  $^4\text{He}$  atoms cannot act as a relaxation mechanism for the  $^3\text{He}$  tunneling reservoir is of the order of a few percent. Let us assume a strain potential between impurity atoms  $i$  and  $j$  of the form  $V_{ij} \sim mc^2(\Delta V/V)^2(a/r_{ij})^3$ , where  $m$  is the mass of the atom,  $c$  is the sound velocity,  $\Delta V/V$  is the volume strain around the impurity atom,  $a$  is the nearest-neighbor distance, and  $r_{ij}$  is the distance between impurities. The tunneling transition of the impurity will then become impossible when the strain energy, of order  $|\nabla V_{ij}|a$ , exceeds the tunneling bandwidth energy,<sup>2</sup>  $\Delta\epsilon \sim 2z\hbar J$ , where  $z$  is the number of nearest neighbors. This is in qualitative agreement with what is observed in  $T_2$  measurements.  $T_2$  decreases as the  $^3\text{He}$  is diluted from  $x = 1$  because the strain field from the  $^4\text{He}$  atoms prevents more and more of the  $^3\text{He}$  atoms from making the  $^3\text{He}$ - $^3\text{He}$  tunneling transitions. As the concentration is decreased below  $10^{-2}$  an increasingly larger number of the  $^3\text{He}$  atoms become free to undergo the  $^3\text{He}$ - $^4\text{He}$  tunneling transition. The impurity atoms are distributed at random so that even in rich mixtures,  $x > 10^{-2}$ , enough of the atoms are apparently in a sufficiently weak strain field to make the tunneling transitions which we have detected in the  $T_1$  experiments.

In summary, we have measured the characteristic impurity tunneling motion  $\tau_{34}$  of  $^3\text{He}$  atoms through the  $^4\text{He}$  lattice by NMR relaxation studies. The volume dependence of the impurity tunneling parameter is much stronger than that of the  $^3\text{He}$  exchange parameter. Both the values of  $T_1$  and  $T_2$  are consistent with the  $\tau_{34}$  which we deduce from the Torrey model.

The authors wish to thank Professor R. A. Guyer and Professor L. H. Nosanow for stimulating discussions of this problem.

†Work supported in part by the National Science Foundation through Grant No. GP-29682 and the Advanced Research Projects Agency through the Cornell Materials Science Center, Report No. 1568.

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