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Nuclear Relaxation of Solid Helium-3 in the bcc Phase*

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We have carried out measurements of the nuclear transverse and longitudinal relaxation times T_2 and T_1 in solid helium-3 between 0.35 and 2°K. Measurements over the whole density range in the bcc phase and for several frequencies between 0.3 and 8.2 Mc/sec are presented. The value of the nuclear exchange parameter J is determined from the T_2 measurements using the theory of exchange narrowing first proposed by Anderson and Weiss. The parameter J is also obtained from the longitudinal relaxation time, which is equal to the relaxation time between Zeeman and the exchange bath over the temperature range where T_1 is temperature-independent. A third determination of J comes from the temperature-dependent T_1 below about 0.6°K, which is interpreted as the exchange lattice relaxation. The values of J from the three different methods are found to agree within experimental error for all the densities investigated. The results are compared with those of the IBM and Oxford workers. A study of the linewidth as a function of frequency shows the expected narrowing as the frequency is increased. To our knowledge, this is the first systematic investigation of the T_2 variation in strongly exchange-narrowed lines that is being reported.

I. INTRODUCTION

SOLID He^3 has some remarkable properties, among which is the unusually large exchange interaction between the nuclear spins. The energy for such an exchange has been expressed in terms of the isotropic Hamiltonian[1]

$$\mathcal{H}=-2J_{12}'\mathbf{I}_1\cdot\mathbf{I}_2\,, \tag{1}$$

where J_{12}' is the exchange integral between two neighboring atoms. Usually J_{12}' is taken to be the same for all nearest neighbors and is neglected for the next-nearest neighbors. This assumption is also made in this paper. Furthermore, in order to conform with recent papers[2,3] on He^3, we write the total exchange Hamiltonian for the system under consideration

$$\mathcal{H}=-\sum_{i>k} hJ\mathbf{I}_i\cdot\mathbf{I}_k\,, \tag{2}$$

where now, J has the dimensions of a frequency.

Measurements of the longitudinal and transverse nuclear relaxation times seem to offer the easiest and most accurate way to measure the coupling J. The first measurements of the relaxation times were published by Goodkind and Fairbank[4] and by Reich.[5] The first authors were able to interpret their results in terms of diffusion relaxation and found an upper limit of the value of J. Reich has published[6] an analysis of his data[5] in terms of exchange and diffusion. More recent work on relaxation times has been carried out by Garwin and Landesman and by Reich *et al.*[3]

The exchange energy corresponds to a Néel temperature $T_N=(z/4)(hJ/k)$, where z is the number of nearest neighbors. T_N varies between about 2 mdeg at the lowest density and about 0.1 mdeg at the highest density of the bcc phase.[2,3] In the hcp phase, T_N decreases further with increasing density.[2,3,6] Direct susceptibility measurements, such as have been performed previously,[7] can only be expected to give reasonably accurate values for T_N if the lowest lattice temperatures attained for the measurement are less than about 20 T_N. These experiments, then are quite feasible with solid He^3 at molar volumes above about 22 cm³. The longitudinal relaxation time T_1, however, becomes exceedingly large at low temperatures, and increases strongly with density.[2,3,8,9] This will prevent reliable susceptibility measurements in the temperature range where T_1 is larger than a few hundred seconds.

After preliminary relaxation measurements, which showed a strong temperature dependence of T_1 at 19.5 cm³/mole[7], it was decided to investigate systematically the relaxation times throughout the bcc phase.

The work to be described has the following objectives: (1) the systematic collection of the values of T_1 and the transverse relaxation time T_2 as a function of frequency at various densities and temperatures in the bcc phase because such a set of measurements has not been published before; (2) the comparison of these relaxation times with the expectations from theory, among them verification of the 10/3 line broadening at

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‡ Alfred P. Sloan Fellow.

[1] See, for example, J. H. Van Vleck, *Theory of Electric and Magnetic Susceptibilities* (Oxford University Press, London, 1932), Chap. 12.

[2] R. L. Garwin and A. Landesman, Phys. Rev. **133**, A1503 (1964).

[3] M. G. Richards, J. Hatton, and R. P. Giffard, Proceedings of the Ninth International Low Temperature Conference 1964 (to be published).

[4] J. M. Goodkind and W. M. Fairbank, in *Helium 3*, edited by J. G. Daunt (Ohio State University Press, Columbus, 1960), p. 52.

[5] H. A. Reich, in *Helium 3*, edited by J. G. Daunt (Ohio State University Press, Columbus, 1960) p. 52.

[6] H. A. Reich, Phys. Rev. **129**, 630 (1963).

[7] A. L. Thomson, H. Meyer, and P. N. Dheer, Phys. Rev. **132**, 1455 (1963).

[8] B. T. Beal, R. P. Giffard, J. Hatton, M. G. Richards, and P. M. Richards, Phys. Rev. Letters **12**, 393 (1964).

[9] R. L. Garwin and H. A. Reich, Phys. Rev. Letters **12**, 354 (1964).

low enough fields; and (3) the derivation of the values of J from both T_1 and T_2 measurements and from the frequency dependence of T_1 in the low-temperature region where T_1 increases sharply with decreasing temperature.

After a review of the current relevant theories of relaxation in Sec. II, a description of the experiment will be given in Sec. III. Finally, in Sec. IV the results and their analysis will be presented.

II. THEORETICAL REVIEW

We shall review here the theories for interpreting the values of the longitudinal and transverse relaxation times T_1 and T_2 which have been proposed in previous publications on solid He^3 so that they can be compared with the experimental results of Sec. IV in a consistent way.

A. Transverse Relaxation

There are two mechanisms which must be considered in the analysis of transverse relaxation: (1) thermally activated diffusion, which is temperature-dependent; and (2) the exchange-modulated dipolar interaction, which is temperature-independent and frequency-dependent. In this work we will be concerned only with the latter. The relaxation by diffusion in solid He^3 has been considered by Goodkind and Fairbank,[4] Reich,[5,6] and Garwin and Landesman.[2]

In solid He^3, for molar volumes greater than about 18 cm^3, $J \gg \gamma H_d$, where H_d is the local dipolar field and γ is the gyromagnetic ratio for He^3, so that the dipolar linewidth is narrowed by the exchange modulation of the spins. Anderson and Weiss[10] have shown that such lines are expected to be Lorentzian except at the "wings" where they become Gaussian. In the limiting case where the Larmor frequency $\omega \gg J$, the value of the transverse relaxation time can be derived from an adiabatic or "truncated" Hamiltonian containing the Zeeman, exchange, and secular dipolar interactions in a rigid lattice. The random-function model of nuclear relaxation,[11] which describes the effects of the exchange coupling as a random modulation of the local dipolar field, predicts that

$$1/T_2 = (\pi/2)^{1/2} M_2^{3/2} M_4^{-1/2}, \tag{3}$$

where M_2 and M_4 are the second and fourth moments of the resonance line. Garwin and Landesman[2] have calculated M_2 and M_4 for the bcc and hcp lattices and use Eq. (3) to find

$$T_2' = \frac{V^2 (J/2\pi) \times 10^{-9}}{17.35} \text{ (sec)} \tag{4}$$

for the bcc lattice, where T_2' refers to the value of T_2 calculated with the adiabatic Hamiltonian, $(J/2\pi)$ is in sec^{-1}, and V is the molar volume in cm^3.

On the other hand, when $J \gg \omega$, the use of the truncated Hamiltonian is no longer valid because the neglected portions of the dipolar interaction then contribute to the broadening of the line. Anderson and Weiss[10] have shown that the line becomes broader by a factor of 10/3 so that

$$T_{2(\omega \ll J)} = \tfrac{3}{10} T_2'. \tag{5}$$

Kubo and Tomita[12] have given a general formulation of the theory of relaxation in which they describe exchange narrowing. They give an expression of the form

$$\Delta\omega_{1/2} = \frac{1}{T_2} = \left(\frac{\pi}{2}\right)^{1/2} \left[\frac{\sigma_0^2}{\omega_{e0}} + \frac{\sigma_{-1}^2 + \sigma_{+1}^2}{\omega_{e1}} \times \exp\left(\frac{-\omega^2}{2\omega_{e1}^2}\right) + \frac{\sigma_2^2}{\omega_{e2}} \exp\left(\frac{-2\omega^2}{\omega_{e2}^2}\right) \right], \tag{6}$$

where σ_k^2 corresponds to the second moment of the absorption line at the frequency $\omega' = (1+k)\omega$, and ω_{ek} corresponds to the frequency of the exchange modulation of the dipolar fields associated with the lines at $\omega' = (1+k)\omega$. The magnitude of ω_{ek} is of the order of the exchange constant J.

For an isotropic or regularly symmetric arrangement of spins Kubo and Tomita show that

$$\sigma_0^2 = \sigma_1^2 = \tfrac{3}{2}\sigma_{-1}^2 = \tfrac{3}{2}\sigma_2^2, \tag{7}$$

and they also assume that $\omega_{e0} = \omega_{e1} = \omega_{e2}$ for the same arrangement. Then, Eq. (6) becomes

$$\frac{1}{T_2} = \left(\frac{\pi}{2}\right)^{1/2} \frac{\sigma_0^2}{\omega_e} \left[1 + \frac{5}{3} \exp\left(\frac{-\omega^2}{2\omega_e^2}\right) + \tfrac{2}{3} \exp\left(\frac{-2\omega^2}{\omega_e^2}\right) \right], \tag{8}$$

with $\omega_e = \omega_{e0}$. In the limit $\omega \gg \omega_e$, Eq. (8) reduces to

$$\frac{1}{T_2'} = \frac{1}{T_{2(\omega \gg \omega_e)}} = \left(\frac{\pi}{2}\right)^{1/2} \frac{\sigma_0^2}{\omega_e}. \tag{9}$$

Identification of Eq. (9) with Eq. (3), the random function model gives $\omega_e = (M_4/M_2)^{1/2} = 2.62J$ where M_2 and M_4 are calculated using equations (3) and (4) of Ref. 2; and Eq. (8) becomes

$$\frac{1}{T_2} = \frac{1}{T_2'} \left[1 + \frac{5}{3} \exp\left(\frac{-\omega^2}{2\omega_e^2}\right) + \tfrac{2}{3} \exp\left(\frac{-2\omega^2}{\omega_e^2}\right) \right], \tag{10}$$

with T_2' defined by Eq. (4). In the limit $\omega \to 0$, Eq. (10) reduces to give the 10/3 ratio of Eq. (5). This theory contains several major approximations; however, the

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[10] P. W. Anderson and P. R. Weiss, Rev. Mod. Phys. **25**, 269 (1953).

[11] A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, New York, 1961), Chap. X.

[12] R. Kubo and K. Tomita, J. Phys. Soc. Japan **9**, 888 (1954).

limiting values in the high and low fields are expected to be accurate.

B. Longitudinal Relaxation

Garwin and Landesman were the first to show[2] that in "pure" He^3 the behavior of T_1 in the solid at low temperatures can be understood in terms of the three-bath model proposed by Bloembergen and Wang.[13] The three energy reservoirs of the model are the Zeeman system, characterized by a temperature T_Z and specific heat $C_Z=(N/4)((\hbar\gamma H)^2/kT_Z^2)$, where N is the number of spins; the exchange system, characterized by a temperature T_E and specific heat $C_E=\frac{3}{32}\times(Nz(\hbar J)^2/kT_E^2)$; and the lattice having a temperature T_L.

The temperature of the Zeeman system can relax to that of the lattice by means of diffusion with a characteristic time τ_{ZL} and to the exchange bath by means of cross relaxation with a characteristic time τ_{ZE}. The temperature of the exchange system can relax to that of the lattice by diffusion, and by phonon interactions with the spins, with a characteristic time τ_{EL}.

There are three temperature regions of interest: (1) the high-temperature region, above about 1°K, in which diffusion of the spins through the lattice is the dominating relaxation process; (2) an intermediate-temperature region, where diffusion becomes negligible and the dominant relaxation process is through the exchange system, which is coupled to the lattice by the modulation of the exchange field; and (3) the low-temperature region where the lattice vibrations decouple the exchange bath from the lattice causing a "bottleneck" for the thermal relaxation of the spins.

The first two regions and the intermediate range between them have been treated theoretically by Hartmann,[14] and by Garwin and Landesman.[2] In region 2, which interests us particularly, Hartmann's expression reduces to

$$\frac{1}{T_1}=\frac{1}{\tau_{ZE}}=\frac{14.5\hbar^2\gamma^4 I(I+1)}{d^2\langle\Delta\omega_1^2\rangle^{1/2}}\times\left\{\exp\frac{-\omega^2}{2\langle\Delta\omega_1^2\rangle}+4\exp\frac{-2\omega^2}{\langle\Delta\omega_1^2\rangle}\right\}, \quad (11)$$

where d is the lattice constant and $\langle\Delta\omega_1^2\rangle$ represents the second moment of the line in zero external applied field, reflecting the coupling between the exchange interaction and the nonsecular part of the dipole interaction. For a bcc lattice, Hartmann[14] finds

$$\langle\Delta\omega_1^2\rangle=7.59J^2I(I+1). \quad (12)$$

Then Eq. (11) can be written as

$$\frac{1}{T_1}=\frac{K}{V^2J}\left\{\exp\left(\frac{-\omega^2}{2\langle\Delta\omega_1^2\rangle}\right)+4\exp\left(\frac{-2\omega^2}{\langle\Delta\omega_1^2\rangle}\right)\right\}, \quad (13)$$

with $\langle\Delta\omega_1^2\rangle^{1/2}=2.38J$, and $K=10.8\times10^{10}$. As $\omega\to0$, one would expect T_1 to become equal to 3/10 T_2'. It can be readily calculated that the limiting value of T_1 from Hartmann's theory is about 30% smaller than the T_2 from Eq. (5).

Garwin and Landesman[2] obtain

$$\frac{1}{T_1}=\frac{1}{\tau_{ZE}}=\frac{(2\pi)^{1/2}}{4}\left[\frac{(\gamma H_d)^2}{\omega_e}\right]\times\left\{\exp\left(\frac{-\omega^2}{2\omega_e^2}\right)+4\exp\left(\frac{-2\omega^2}{\omega_e^2}\right)\right\}, \quad (14)$$

where H_d is a phenomenological constant to be associated with the local dipolar field. They derive the relation $(\gamma H_d)^2\approx2.2M_2$ to evaluate γH_d, and then determine ω_e by equating their relation for T_1 as $\omega\to0$ with that for $T_2'\times3/10$ giving

$$\omega_e=1.76J[zI(I+1)]^{1/2}, \quad (15)$$

and when Eq. (14) is reduced to the form of Eq. (13), the value of the constants become $\omega_e=4.30J$ and $K=7.23\times10^{10}$. These values disagree significantly with those obtained by Hartmann.

In our own analysis of the T_1 data, which will be described later, we make use only of the asymptotic behavior of T_1 as $\omega\to0$. Discrepancies in the exact relationship between J and ω_e will not be labored further because, as will be seen in Sec. IV, the exponential form of Eq. (13) can not be made to fit the observed values of T_1 regardless of the value of ω_e.

In region 3 the thermal relaxation of the Zeeman bath to the lattice temperature by way of the exchange bath can be described by the set of coupled differential equations[15]:

$$\frac{d}{dt}\left(\frac{p}{T_Z}\right)=\frac{p}{\tau_{ZE}}\left(\frac{1}{T_E}-\frac{1}{T_Z}\right)=0 \quad (16a)$$

and

$$\frac{d}{dt}\left(\frac{q}{T_E}\right)=\frac{q}{\tau_{EL}}\left(\frac{1}{T_L}-\frac{1}{T_E}\right)+\frac{p}{\tau_{ZE}}\left(\frac{1}{T_Z}-\frac{1}{T_E}\right) \quad (16b)$$

where $p=\frac{1}{4}N((\hbar\gamma J)^2/k)$ and $q=(3N_Z/32)(\hbar J)^2/k$.

If the conditions $\tau_{ZE}\ll\tau_{EL}$, snf $C_Z/C_E\lesssim10$ exist, the asymptotic behavior of T_Z at times $t\lesssim\tau_{LE}$ and $t\gtrsim5\tau_{ZE}$ can easily be seen: Initially we let rf pulses of the appropriate resonant frequency ω saturate the Zeeman system at a sufficient rate and for a sufficient time that the heat flowing out of the Zeeman bath elevates the

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[13] N. Bloembergen and S. Wang, Phys. Rev. **93**, 72 (1954).

[14] S. R. Hartmann, Phys. Rev. **133**, A17 (1964).

[15] R. B. Griffiths, Phys. Rev. **124**, 1023 (1961).

temperature of the exchange bath above that of the lattice. Then $T_Z \gg T_E > T_L$. After the pulses have been cut off, Eq. (16a) describes the initial decrease of the Zeeman temperature toward that of the exchange bath with a characteristic time τ_{ZE}. After a time $t_1 \gtrsim 5\tau_{ZE}$ the Zeeman temperature has fallen approximately to that of the exchange system, and the rate at which both reservoirs approach the temperature of the lattice can be described approximately by the addition of Eqs. (16a) and (16b) with $T_E \simeq T_Z$ so that

$$\left(1+\frac{C_Z}{C_E}\right)\frac{d}{dt}\left(\frac{1}{T_Z}\right)=\frac{1}{\tau_{EL}}\left(\frac{1}{T_L}-\frac{1}{T_Z}\right). \tag{17}$$

Then

$$\frac{1}{T_Z}=\left(\frac{1}{T_Z}-\frac{1}{T_L}\right)_{t=t_1}\exp\left(-t\left\{\left[1+\frac{C_Z}{C_E}\right]\tau_{EL}\right\}^{-1}\right)+\frac{1}{T_L}. \tag{18}$$

We also define $T_1^* \equiv [1+C_Z/C_E]\tau_{EL}$.

Griffiths[15] has analyzed the single- and double-phonon relaxation precesses, first proposed by Waller,[16] for the exchange lattice relaxation. He finds that the 2-phonon, or Raman, process is the most significant except at very low temperatures, where the one-phonon process predominates. Garwin and Landesman[2] have applied Griffiths' result to the case of solid He³ and show that for $T \ll \Theta_D$ the Debye temperature, the exchange-lattice relaxation due to the two phonon varies with temperature as T^{-7}. By applying the result of Garwin and Landesman to the bcc phase we obtain

$$\tau_{EL}^{-1}=2.56\times10^{-35}\left(\frac{d^2J}{da^2}\right)^2\frac{T^7}{\Theta_D^{10}}, \tag{19}$$

where a is the nearest-neighbor distance.[17]

Recently, Richards[18] has made a calculation in which he has extended the theory of Griffiths to include the effects of the correlations due to exchange between neighboring spins, which were neglected by Griffiths. As a result of the spin correlations, he finds that in the bcc lattice τ_{EL}^{-1} should be about 15% less than the value Griffiths has given.

C. Summary

In conclusion, it may be stated that there exist approximate theories relating the longitudinal and transverse relaxation with the exchange parameter J and the resonant frequency ω. It can at least be expected that general expressions exist of the form

$$T_2=\tfrac{3}{10}T_2'\Phi(\omega/J)\,,\quad\text{where}\quad 1\leqslant\Phi(\omega/J)\leqslant 10/3 \tag{20a}$$

and

$$T_1=\tfrac{3}{10}T_2'\Psi(\omega/J)\,,\quad\text{where}\quad 1\leqslant\Psi(\omega/J)<\infty\,. \tag{20b}$$

In systems with electronic paramagnetism, where there is strong exchange narrowing, one usually has $\omega \ll J$, even for magnetic fields as high as say 50 kG. He³, as we shall see, is rather exceptional because by varying the density, J can be changed in such a way that the variation of the linewidth by a factor of approximately 10/3 as a function of frequency can readily be observed.

III. EXPERIMENTAL APPARATUS AND PROCEDURE

A. The Cryostat

At the beginning of this research, it appeared of interest to carry out relaxation measurements at pressures up to 15 000 psi and a new cavity for this high-pressure range was designed and constructed, partly based on drawings by Garwin kindly given to us by Reich. The chief differences are that our high pressure seals are made of Araldite, while the IBM workers have used nylon seals, and in our design the He³ sample is packed inside the rf coil. In addition, there is a manganin pressure gauge[7] inside our cavity about 7 mm above the top of the rf coil. A copper rod in contact with a He³ refrigerator penetrates the high-pressure cell and is connected to thin copper wires embedded in the Araldite which surrounds the He³ sample. In this way good thermal contact is established between the He³ bath and the sample. The "pure" He³ used in the experiment contained less than 0.04% He⁴.

The temperature was measured by a 450 Ω Speer carbon resistor, the resistance of which is practically independent of magnetic field in the temperature range used. The carbon resistance thermometer was calibrated between 0.8 and 1.5°K by means of the He³ vapor pressure. Below 0.7°K, it was calibrated against the amplitude of the nuclear resonance signal of solid He³ with a density of about 22.5 cm³/mole. This amplitude which was measured with free-precession techniques, is known to follow Curie's Law[7] within the limits of experimental error. Hence the absolute temperature was known to within about 8 mdeg below 0.7°K, and to better than about 3 mdeg above this temperature, an accuracy considered adequate for the type of measurements planned.

B. The Pressure- and Vacuum-Generating System

We used the blocked-capillary technique for forming the solid He³ in the cavity. This technique and the way in which the manganin pressure gauge was employed to evaluate the density of the solid, are the same as that

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[16] I. Waller, Z. Physik **79**, 370 (1932).

[17] In obtaining Eq. (19), Garwin and Landesman's expression for τ_{EL}^{-1} has been multiplied by 3 to account for the error in transcribing Griffiths' equation, as pointed out by P. M. Richards in Ref. 18.

[18] P. M. Richards, Phys. Rev. **137**, A1327 (1965).

discussed in a previous paper.[7] The stainless-steel capillary which passed through the He^4 "pot" to the sample cavity had an i.d. of 0.020 in. but was filled with a manganin wire with a diameter of 0.018 in. to decrease the cross-sectional area of the capillary so that it would block more readily. The pressure and vacuum generating equipment are also essentially the same as that used previously.[7]

In forming solids with molar volumes greater than about 23 cm^3 we found that the pressure in the solid, which we measured with the manganin gauge, was always greater than that which was predicted by the use of P-T and V-T[19] diagrams with the assumption that the capillary blocked when the melting curve was reached from the high-temperature side. The pressure used in the liquid to form the sample with a molar volume of 24.6 cm^3 was only 0.5 atm greater than the final pressure measured in the solid. The exact pressure finally found in the solid depended upon the rate of cooling of the sample cavity by the He^3 refrigerator, and could only be known by the values measured with the manganin pressure gauge.

C. The Electronic Equipment

The components of the electronic system used in the experiment are: (1) a multiple-pulse trigger with variable delay times; (2) two Tektronix Type 163 pulse generators; (3) a General Radio Type 1001 A standard signal generator; (4) an rf gate circuit,[20] constructed after the design of Blume; (5) a wide-band rf amplifier and detector; and, (6) a Tektronix Type 564 storage oscilloscope.

The tank circuit consisted of a coil, 1.2 cm long and 0.30 cm i.d., wound with 65 turns of number 36 copper wire, and a variable capacitor which was located on top of the cryostat. The coil was first filled with epoxy and then reamed out inside so that the filling factor of the sample He^3 was about $\frac{1}{2}$.

The coherent-pulse technique, described elsewhere in the literature of NMR,[21] was employed to detect the nuclear signal in measurements of T_2 and of T_1 for values less than about 0.5 sec. A combination of cw method and pulse method was used to measure values of T_1 longer than 0.5 sec.

In order to measure T_2, the exponential envelope generated by echoes following the pulse sequence 90–180°, with different delay times, was observed on the storage screen of the oscilloscope. Since the scope was triggered by the 90° pulse, T_2 was the mean characteristic decay time determined from the plot of $\ln H_{\text{echo}}$ versus time, where H_{echo} is the amplitude of the echo measured on the oscilloscope. The envelope was left on the storage screen long enough for the values to be read, and T_2 to be determined, before the picture was erased. The envelopes were always found to be exponential.

To measure values of T_1 less than 0.5 sec, an envelope of signals following 90° pulses at different times t after an initial 180° pulse at $t=0$ was traced out on the storage screen. The envelope was the curve $H=H_0(1-2e^{-t/T_1})$ so that T_1 also could be immediately determined from the mean decay time in a plot of $\ln(H_0-H)$ versus t. In the cases where single relaxation times were observed, it was usually found that sufficient accuracy could be obtained by locating the null in the envelope at time t_N where one has $T_1=t_N/\ln 2$. If two relaxation times were apparent, the full envelope was analyzed.

Since it was necessary to wait a period of at least $5T_1$ between successive pulse sequences, this method became rather cumbersome for measuring relaxation times longer than about 0.5 sec. Instead, a cw method using a Rollins circuit was employed, and a small 10-cps magnetic field modulated the dc magnetic field. One or more 90° pulses were applied to saturate the signal and the oscilloscope was triggered by the last pulse. The recovery of the signal as a function of time was then recorded on the oscilloscope.

IV. RESULTS

The relaxation times were measured at frequencies between 0.3 and 8.2 Mc/sec at nine molar volumes between 20.1 and 24.6 cm^3. The values of T_2 and T_1 over

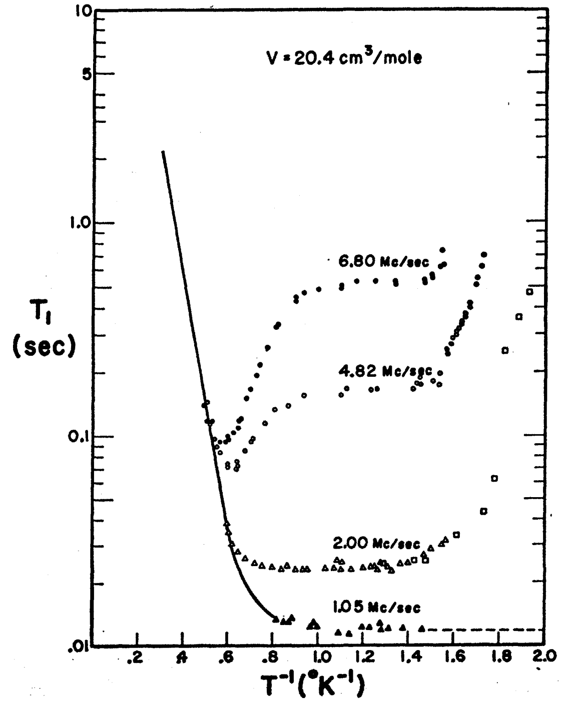

FIG. 1. The temperature variation of T_1 for several frequencies as a function of T^{-1} for the molar volume of 20.4 cm^3.

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[19] E. R. Grilly and R. L. Mills, Ann. Phys. (N. Y.) 8, 1 (1959).
[20] R. J. Blume, Rev. Sci. Instr. 32, 554 (1961).
[21] A. Abragam, Ref. 9, Chap. III, for example.

TABLE I. The transverse relaxation time T_2 in milliseconds for several frequencies (inMc/sec) at the given molar volume in the bcc phase.

$V \backslash \omega/2\pi=$0.53		0.69	1.05	1.59	2.00	3.56	4.82	6.80	8.20
20.1	6.5±1.5		7.5±0.7		9.4±1	13.6±1	15.4±0.7	15.7±0.7	16.6±0.7
20.4	7.3		9.2					22	
20.5					13		16.5		
21.3	20		24		28		37	39	
21.8	41		42		45		54	56	
22.1		52	55	55	56		63	69	74
22.4	58		66		64		74	74	
24.1	195± 50		200±40		200±30		220±20	210±20	
24.6	240±100		260±80		310±50		300±50	310±50	

The longitudinal relaxation time T_1 in milliseconds for several frequencies (in Mc/sec) at the given molar volume in the bcc phase.

$V \backslash \omega/2\pi=$0.33		0.47	0.53	0.69	1.05	1.59	2.00	3.56	4.82	5.50	6.80	8.20
20.1		7.2±1	7.4±1		11±1		25±2	89±5	220±10		760±40	1700±80
20.4			7.6		11		23				520	
20.5									160			
21.3			20		27		33		64		100	
21.8			41		45		54		79		100	
22.1	50± 5		48	53	56	61	63		84		105	120
22.4			53		67		72		96		115	
23.8	185±20		195	205	215		235		255		270	270
24.1			215		250		270		300		300	
24.6			400±60		440±40		450±40		480±30	490±30	490±30	

the temperature range where they are temperature-independent (plateau region) are contained in Table I. They represent the average of several experimental points at different temperatures in the "plateau" region. The temperature variation of T_1 and T_2 at several frequencies for the molar volume 20.4 cm^3 is shown in Figs. 1 and 2. The frequency dependence of T_1 and T_2 in the "plateau region" is shown in Fig. 3 for three representative densities.

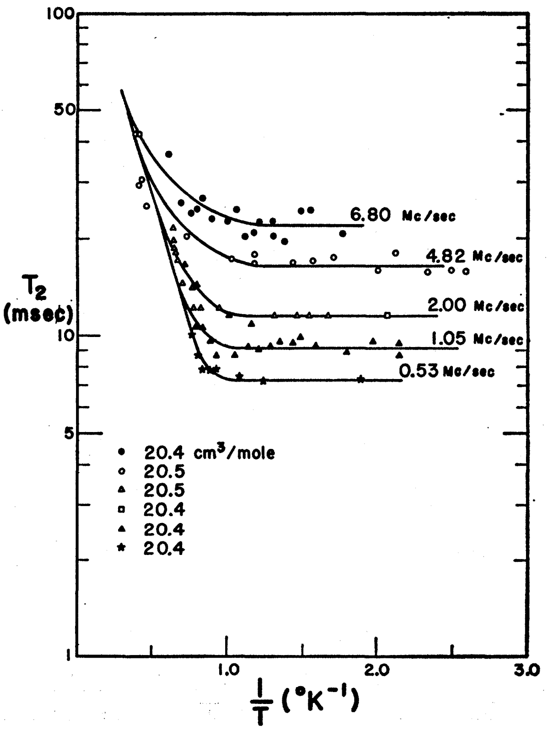

FIG. 2. The temperature variation of T_2 for several frequencies as a function of T^{-1} for the molar volume of 20.4 cm^3.

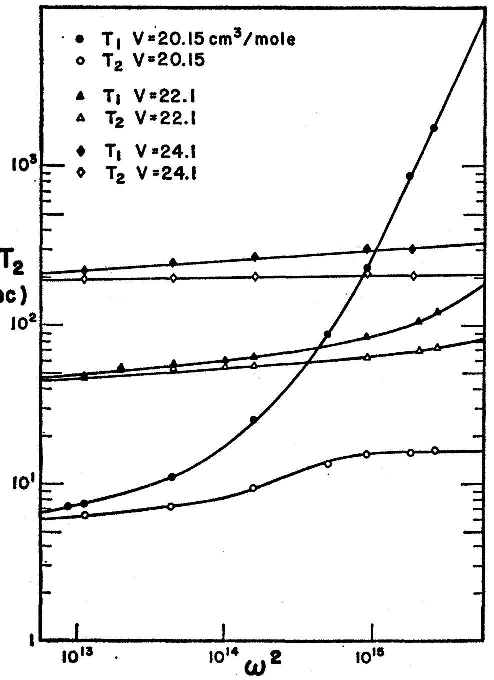

FIG. 3. The frequency dependence of T_1 and T_2 for three representative densities as a function of ω^2 on a logarithmic plot. At the molar volume 20.1 cm^3, the frequency range covered is large enough to show clearly the tendency towards 10/3 narrowing of the linewidth with frequency.

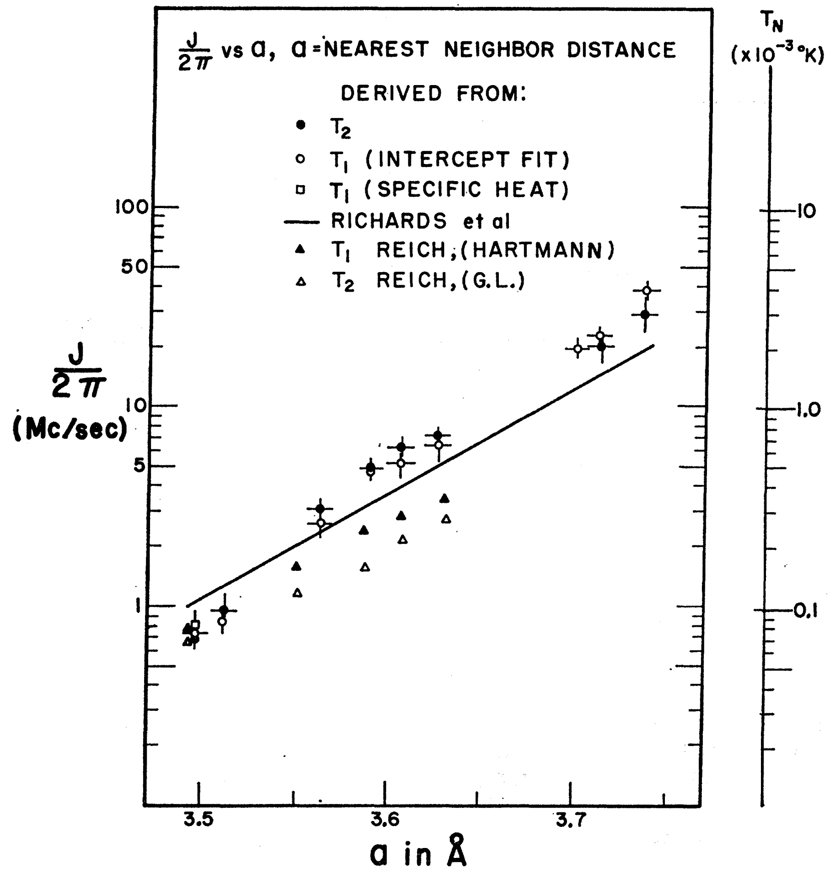

FIG. 4. The exchange parameter $J/2\pi$ in Mc versus the distance a between the nearest neighbors in the bcc phase. The solid line is the average of the results obtained by Richards *et al.* A value of $J/2\pi$ of 10 Mc corresponds to $T_N = 0.95 \times 10^{-3}$°K according to the equation given in the text. For sake of clarity, the theoretical curves of E. M. Saunders [Phys. Rev. **126**, 1724 (1962)] and of L. H. Nosanov and W. J. Mullin [Phys. Rev. Letters **14**, 133 (1965)] have been omitted here.

A. Transverse Relaxation

The values of T_2 contained in Table I were fitted to a common reduced curve as suggested by Eq. (20a). The reduced experimental values of T_2 are shown in Fig. 5. The following procedure was used to obtain the curve shown:

(1) The value of J for the most dense sample was determined from the high-frequency values of T_2, where $\omega \simeq 10J$, so that the adiabatic condition necessary for the use of Eq. (4) prevails. The reduced values of T_2/T_2', where T_2' is the value defined by Eq. (4), were plotted against $(\omega/J)^2$ on a log-log scale.

(2) For each progressively less dense sample, the value of J was varied to give the best fit of the reduced data to the curve previously generated.

The values of J used to obtain the best fit at each density appear in Fig. 4. Although the errors in such a procedure accumulate, it is felt that there is sufficient overlap between the reduced data for each density for the error in J at the lowest density to be less than 10%.

The probable error in the value of T_2 for each frequency is represented in Fig. 5 for the data obtained at 20.1 cm^3. The errors for the same frequencies at other densities are the same, except at the lowest density. At the lowest density, the experimental difficulties encountered in measuring such long values of T_2 caused the probable error to be greater at each frequency by a factor of 3. In fitting the reduced data, the values of T_2 obtained at 0.5 Mc/sec were given the least weight because at that frequency the signal to noise ratio had become so unfavorable that the probable error is 20%.

The solid line in Fig. 5 represents the behavior of T_2 with frequency predicted by Kubo and Tomita. The values of T_2 measured tend to decrease to the zero-field value less rapidly than predicted by Eq. (10), but agree with the theory in the limiting cases $\omega/J \rightarrow 0$ and $\omega/J > 10$.

The values of J determined by Garwin and Landesman from Reich's measurements of T_2 appear in Fig. 4. Interpolation of our data to the frequency used by Reich produces values of T_2 in agreement with his data for all densities. The increasingly large discrepancy between Reich's results for J and ours as the density is decreased is caused by the line broadening which was not considered in the calculation of Garwin and Landesman.

B. Longitudinal Relaxation

The values of T_1 contained in Table I were also fitted to a reduced curve as shown in Fig. 6. The procedure used to generate the function suggested by Eq. (18b) is similar to the one used for T_2:

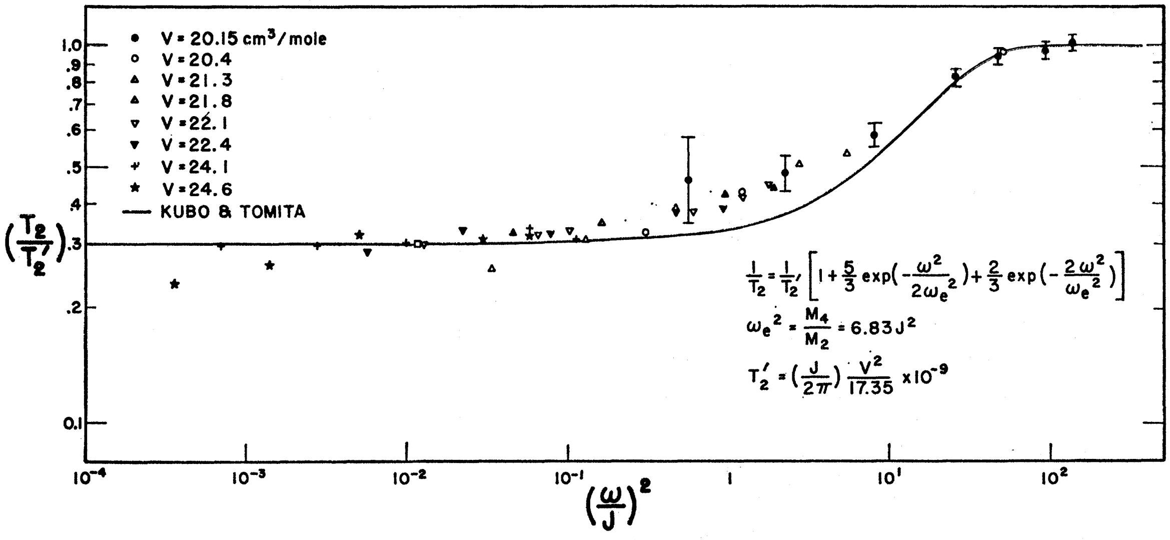

FIG. 5. The reduced transverse relaxation time T_2/T_2' versus the reduced frequency $(\omega/J)^2$. The solid line is Eq. (8) derived from the calculations of Kubo and Tomita.

(1) An arbitrary value of $J(=J_0)$ was assumed for the highest density sample, and the reduced values $T_1/(J_0V^2)$ were plotted against $(\omega/J_0)^2$ on a log-log scale.

(2) As in the T_2 procedure, J_0 was varied for each less dense sample until the reduced values of T_1/J_0V^2 obtained gave the best fit to curve generated by the values at higher densities.

(3) The reduced curve generated by this procedure gives the extrapolation of the value of T_1 of the highest density used at $\omega\rightarrow0$. Then, since $T_{1(\omega\rightarrow0)}=T_{2(\omega\rightarrow0)}=(3/10)T_2'$, the value of J for that density was calculated from Eq. (4), and the multiplicative factor B, was determined so that $J=BJ_0$. The values of J for all other densities were determined by multiplying the values of J_0 obtained in the fitting procedure by the same factor. These values also appear in Fig. 5.

It is felt that such an extrapolation is valid because for all densities in the bcc phase, $J\gg\gamma H_d$, so that the chief effect of decreasing the density is to vary the range of values of T_1 that can be experimentally observed as a function of (ω/J).

The accumulated error in the procedure above is again felt to be less than 10%. The extrapolated values of T_1 and T_2 in zero field for the highest density, produced by the fitting process, agree within 5%; and there is no systematic disagreement between the values

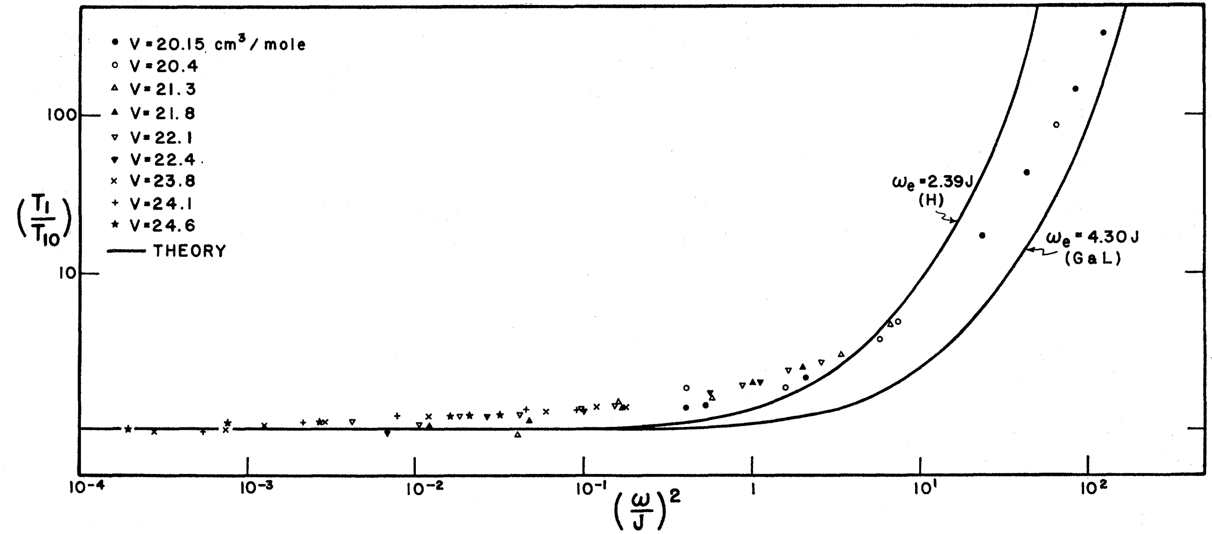

FIG. 6. The reduced longitudinal relaxation time T_1/T_{10} versus the reduced frequency $(\omega/J)^2$. For the experimental points T_{10} has been taken as indicated in the text. The solid curves are the reduced theoretical relaxation times (Refs. 4 and 13). For Hartmann's curve, T_{10} was taken as the expression before the brackets in Eq. (11). For Garwin and Landesman's curve, T_{10} was taken as $\frac{3}{10}T_2'$.

of J from the two sources, T_1 and T_2 at any density. The experimental values of T_1 and T_2 in the lowest fields used were found to be the same within their experimental errors at all densities.

The behavior of T_1 with ω predicted by Hartmann, and by Garwin and Landesman is also indicated in Fig. 6. Both equations were normalized to fit the reduced data in zero field. It is apparent that the form of the two exponential frequency terms will not fit the reduced data regardless of the relation selected between J and ω_e.

The values of J calculated by Hartmann for the T_1 data of Reich also appear in Fig. 4. The interpolation of our T_1 data to 5.2 Mc/sec gives the same values as those of Reich, and therefore the discrepancy arises only as a result of the way in which J is calculated.

The values of J determined from T_1 and exchange-bath specific-heat measurements by Richards *et al.*[3] appear in Fig. 4 also. Although our results agree with theirs within the combined experimental errors, the slope of the dependence of $\ln J$ upon a, the nearest-neighbor distance, is larger in our measurements. This can partially be explained by the fact that they determined J from their T_1 measurements by fitting their data at the higher densities to the exponential behavior of the equation by Hartmann.[22]

For the molar volume of 20.15 cm^3, a careful study was made of the longer component of the longitudinal relaxation ($\equiv T_1{}^*$). This study was to determine J from the specific heat of the exchange bath and to determine the temperature dependence of τ_{EL}.

The values of $T_1{}^*$ versus $(1/T)$ at this density are presented in Fig. 7 on a logarithmic scale for three different frequencies. By plotting $T_1{}^*$ versus ω^2 at several temperatures, straight lines were obtained with a common negative intercept on the ω^2 axis. Since the relaxation observed is $T_1{}^*=(1+R)\tau_{EL}$ where $R=C_z/C_e=\omega^2/(3J^2)$, the magnitude of the intercept is $3J^2$. The value of J determined from this procedure is in satisfactory agreement with that of the other methods and also appears in Fig. 4.

The magnitude of τ_{EL} measured is in satisfactory agreement with the predictions of Eq. (19) when the effects of atomic diffusion are considered in the calculation,[23] and the temperature dependence is in qualitative agreement with that of other workers.[8] The order of magnitude of τ_{EL} is also consistent with that previously reported[7] by this laboratory at a volume 19.5 cm^3 and frequency of 3 Mc/sec.

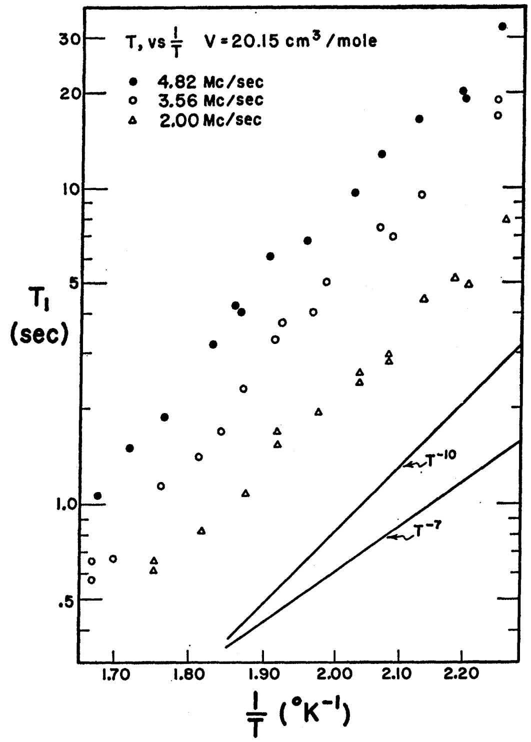

FIG. 7. The exchange-lattice relaxation time as a function of T^{-1} for three different frequencies (log-log scale).

C. Effect of He^4 Impurities

A series of experiments was also made on He^3 containing an impurity of 0.5% He^4. This study was of interest since the effect of small quantities of He^4 had been most important in previous experiments,[24,7] introducing apparent departures from Curie's law and

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[22] The magnitudes of T_1 measured by Richards *et al.* (communicated by M. G. Richards to the authors) at the molar volumes of 23 and 24 cm^3 are about 30% less than the values we obtain from interpolation of our data to the same density and frequency. Since our data is in much better agreement with theirs at the higher densities, we suggest that their use of the blocked-capillary technique to determine the molar volume of the solid introduces an uncertainty in the exact pressure in the solid at lower densities. The discrepancy in the magnitude of T_1 can be resolved if one assumes that the plug in their capillary slipped enough while forming the less dense solids to decrease the molar volume by about 0.5 cm^3.

[23] Garwin and Landesman in Ref. 2 first pointed out that the exchange lattice relaxation time due to diffusion is of the order of τ_c, the characteristic jump time of the atomic diffusion. P. M. Richards has shown in Ref. 18 that $\tau_{EL}{}^{-1}$ due to diffusion is given by $2(z-1)/\tau_c$. By using the relation $\tau_c{}^{-1}=6D/a^2$, where D is the diffusion-coefficient data given by Reich in Ref. 6 for $V=20.12$ cm^3 ($D=4.3\times10^{-5}\times\exp(-13.6/T)$ cm^2/sec), we find that at $T^{-1}=2.1$°K^{-1} the exchange lattice relaxation time due to diffusion is 0.77 sec. By using Eq. (19) and the 15% correction factor due to exchange correlations we calculate τ_{EL} due to the two-phonon process to be 6.3 sec at the same temperature. We use for d^2J/da^2 at this molar volume the value $(1.74\pm1.0)\times10^3$ Mc/sec Å^{-2}, obtained graphically from our values of J in Fig. 5. Then, the "theoretical" value of T_1 at this temperature for the frequency 4.72 Mc which is predicted by these calculations is $T_1{}^*=(1+R)\{\tau_{EL(\mathrm{diff})}{}^{-1}+\tau_{EL(\mathrm{phonons})}{}^{-1}\}^{-1}=9.0$ sec. The value which was measured is 12 sec. At 2.25°K^{-1}, where we measure $T_1{}^*=33$ sec, the value calculated by the same process is 43 sec; and at 1.7°K^{-1}, where $T_1{}^*$ measured 1 sec, the calculated value is 0.044 sec. Therefore, even though the magnitude of τ_{EL} which we measure is consistent with the theory over most of the temperature range (within the error of evaluating d^2J/da^2), τ_{EL} increases less rapidly with decreasing temperature than the theory would predict in this interval.

[24] E. D. Adams, H. Meyer, and W. M. Fairbank, *Helium 3* (Ohio State University Press, Columbus, 1960), p. 57.

inconsistencies in the values of the exchange interaction derived from different methods. Our conclusions are, that for a given density, neither T_2 nor τ_{ZE} are appreciably affected by the impurity. On the other hand, the relaxation process at lower temperatures, where the long τ_{EL} causes a "bottleneck," is very much affected. The exchange specific heat seems to have increased by a large amount and it becomes more difficult to heat up the exchange system. No quantitative conclusion could be made from our data. It appears possible as suggested by Richards *et al.*[3] that the 3-bath model is no longer adequate and that there might be a fourth bath situated between the exchange system and the lattice. Only further systematic experiments with very dilute solutions of He^3 in He^4 can help to clarify this matter.

V. CONCLUSION

By a systematic study of the frequency dependence of T_1 and T_2 in "pure" He^3 we have been able to obtain normalized empirical relations of T_1 and T_2 versus (ω/J) valid for all the different values of the exchange interaction. Therefore, we were able to derive in a consistent way this exchange constant from both types of measurements. We have been able to verify by this procedure the 10/3 broadening of the linewidth when $J \gg \omega$. To our knowledge such a systematic investigation by NMR[25] or ESR[26] has not yet been published for any other compound. Effects of He^4 impurities on the relaxation times are already important if the He^4 concentration is 0.5%. Further relaxation studies with small (0.1 to 0.3%) He^4 doping are planned, in order to get a better understanding of the relaxation mechanisms.

ACKNOWLEDGMENTS

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[25] *Note added in proof.* R. N. Rogers, J. G. Pribram and P. D. Lazay, [Bull. Am. Phys. Soc. 9, 740 (1964)] have recently presented results on the exchange narrowing in EPR measurements in $Cu(NH_3)_4SO_4 \cdot H_2O$.

[26] *Note added in proof.* We have been informed by Dr. R. L. Garwin and Dr. H. A. Reich that they have measured before our own experiments the frequency dependence of T_1 and T_2 at about 19.5 cm^3/mole in the hcp phase in solid He^3. Their unpublished measurements show the expected increase of T_2 with frequency. Our own more recent measurements at the same density at 14 different frequencies between 0.36 and 4.5 Mc/sec show that the frequency variation of T_2 is in excellent agreement with the theory of Kubo and Tomita. In the bcc phase on the other hand, the values of T_2 in the middle frequency region tend to lie above the theoretical curve as seen in Fig. 5.