

A10.2 NMR INVESTIGATION OF BCC SOLID  $\text{He}^3$ \*. Wijit Senghaphan\*\* and George O. Zimmerman. Physics Department, Boston University, Boston, Massachusetts.

The application of nuclear magnetic resonance techniques to the investigation of solid  $\text{He}^3$  has been particularly successful in exploring the nature of the spin-spin and the spin-lattice interaction. In this paper we would like to report some results and interpretations which resulted from our low temperature NMR investigation of BCC  $\text{He}^3$ . First we offer an alternate explanation for the long spin lattice relaxation time below about  $0.2^\circ\text{K}$  observed by us and others<sup>1,2</sup> in the BCC phase. Second, we report the measurement of the exchange interaction,  $J$ , by a new method. Third, we report preliminary results of the  $J$  dependence of phonon absorption by the spin system. Fourth, we offer some observations on the importance of next nearest neighbors in BCC solid  $\text{He}^3$ .

The long spin lattice relaxation times below  $0.2^\circ\text{K}$  observed by Richards et al<sup>1</sup>, us, and investigated by Hunt et al<sup>2</sup> can be described by the solution to the rate equations.

$$\begin{aligned} 1) \quad \frac{C_z}{k} \frac{dT_z}{dt} &= k_z \dot{\beta}_z = -\eta_1 k_z (\beta_z - \beta_x) \\ 2) \quad \frac{C_x}{k} \frac{dT_x}{dt} &= k_x \dot{\beta}_x = -\eta_2 k_x (\beta_x - \beta_\ell) + \eta_1 k_z (\beta_z - \beta_x) \\ 3) \quad \frac{C_\ell}{k} \frac{dT_\ell}{dt} &= C_\ell T_\ell^2 \dot{\beta}_\ell = -\eta_3 C_\ell T_\ell^2 (\beta_\ell - \beta_B) + \eta_2 k_x (\beta_x - \beta_\ell) \end{aligned}$$

where  $\beta = \frac{1}{kT}$ ,  $C$  denotes the specific heat,  $T$  the absolute temperature, the subscripts  $z$ ,  $x$ ,  $\ell$  and  $B$  refer to the Zeeman, exchange, lattice and bath reservoirs respectively.  $k_z = T_z^2 C_z$ ,  $k_x = T_x^2 C_x$  and the  $\eta$ 's are characteristic coefficients where  $\eta_1 = (T_{zx})^{-1}$  of Richards et al<sup>1</sup>. Equations 1 and 2 are the rate

equations of Richards et al<sup>1</sup> in the Nosanow-Varma<sup>3</sup> notation, equation 3 describes the heat flow between the lattice and bath. At times long compared with the Zeeman-exchange relaxation time we can assume that  $\beta_x = \beta_z$  and the approximate solution becomes

$$4) \beta_z(t) = \beta_z^\infty + c'e^{-\lambda'_1 t} + d'e^{-\lambda'_2 t}$$

$$\text{where } \lambda'_1 = \frac{(1 + \rho')}{1 + \rho} \eta_2 \quad \lambda'_2 = \frac{\eta_3}{1 + \rho}$$

$$5) \rho = \frac{k_x}{k_z} \quad \text{and} \quad \rho' = \frac{k_x + k_z}{c_\ell T_\ell^2} = \frac{k_x + k_z}{k_\ell T_\ell^5} \quad \text{with}$$

$$k_\ell = \frac{C_\ell}{T_\ell^3}$$

6)  $\eta_2 = 1/T_{x\ell}$  where at low temperatures  $T_{x\ell}$  is dominated by the temperature independent exchange diffusion<sup>4,5</sup> and can be shown to be

7)  $T_{x\ell} = \kappa \left[ \frac{2(\rho + 1)}{9J} \right]$  which has the same J dependence as  $\gamma_D$  in Eq. 1 of Reference 2.  $\eta_3$  determines the lattice bath heat flow which at low temperature is limited by the boundary resistance, i.e.

8)  $\eta_3 = \frac{A}{RC_\ell} = \frac{1}{T_{\ell B}}$  where A is the contact area between the bath

and the lattice and R is the boundary resistance which is proportional to  $T^{-3}$ .

We believe that the  $J^{-1}$  dependence of the characteristic times observed by Hunt et al<sup>2</sup> is due to the predominance of  $\eta_2$  in chambers 2 and 3 while the relaxation in chamber 1 was limited by  $\eta_3$ . The latter was also the case with our observations of the low temperature relaxation time at  $V = 22.1 \text{ cm}^3$ . Assuming  $T_{x\ell} = 4.5 \text{ sec}$  as measured, and  $T_{\ell B} = 180 \text{ sec}$ . we obtain  $R \approx \frac{5 \times 10^4}{T^3}$  deg  $\text{cm}^2/\text{watt}$ , in fairly good agreement with the values obtained by others<sup>6</sup>.  $C_\ell$  was evaluated from the measurements of Sample and

Swenson.<sup>7</sup>

Varma<sup>8</sup> suggested that the exchange interaction J can be measured by comparing the values of  $\tau_0$ , the time in which the Zeeman reservoir reaches infinite temperature after the application of a  $180^\circ$  pulse, at high,  $\tau_0^h$ , and at low,  $\tau_0^l$ , temperatures. At high temperatures the exchange reservoir is closely coupled to the lattice. At low temperatures it is decoupled from the lattice and is consequently heated by the energy from the Zeeman reservoir.  $C_x$  can be determined from the relation

$$9) \quad \tau_0^l / \tau_0^h = \left(1 + \frac{C_z}{C_x}\right)^{-1} \left[ \frac{1 - \ln\left(1 - \frac{C_z}{C_x}\right)}{\ln 2} \right] \quad \text{for } \frac{C_z}{C_x} < 1.; \quad \frac{C_z}{C_x} = \frac{\omega_0^2}{3J^2}$$

Since the accuracy of this determination of J is greatest when  $.9 < \frac{C_z}{C_x} < 1$  our determination of J was limited by

the highest NMR frequency we could achieve in our apparatus which was 10.25 Mc/sec. The values of J measured in this way are given in Table 1. The values are in good agreement with those of others and generally fall between those of Richardson et al<sup>9</sup> and Panczyk et al<sup>10</sup>.

TABLE 1

$V(\text{cm}^3/\text{mole})$	$J/2\gamma$ (mc/sec)
22.05	5.25
22.10	5.02
22.4	5.7
22.7	6.6

In a previous publication<sup>11</sup> we reported the partial saturation of the NMR signal on application of a 9.91-Mc/sec sound wave to the sample. This indicated that the spins absorbed energy from the low frequency, 9.91-Mc/sec. phonons. Although the application of a sound signal increased the spin lattice relaxation time, no dependence of this increase on J or the NMR frequency was observed. Further investigation, however, suggests

that the partial saturation at a particular ultrasonic energy is dependent on  $J$ . Figure 1 shows the partial saturation of the NMR signal upon the application of sound pulses, having the same energy, to a previously unsaturated sample. The saturation was observed below  $0.2^{\circ}\text{K}$  where the spin lattice relaxation time is constant. The energy of the sound signal was assumed to be proportional to  $V^2t$  where  $V$  is the voltage applied to the ultrasonic transducer and  $t$  is the duration of the signal. The points suggest a broad maximum in the absorption, with the maximum at  $J$  equal to the ultrasonic frequency of 9.91 Mc/sec. The broken line shows the suggested saturation curve. It must be emphasized

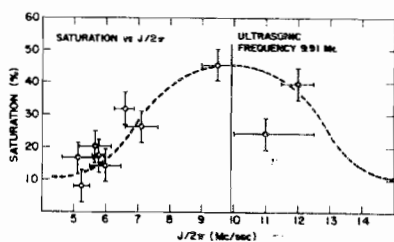


Figure 1

that the data points are preliminary. The errors in  $J$  are caused by the uncertainty in the blocking pressure of the capillary and in  $J$  itself, although at  $J < 8$  Mc/sec. we could measure  $J$  by the method

reported earlier in this paper. In that case  $J$  was measured in the same experimental runs as the absorption. The uncertainty in saturation is brought about by the imperfect coupling of the transducer to solid  $\text{He}^3$ . Further investigations of this effect are in progress and will be extended to lower ultrasonic frequencies.

Experimental low temperature investigations of solid  $\text{He}^3$  have been capable of measuring  $J$  and also observing the effects of its energy spectrum. Instances of the latter are phonon energy absorption by the spins, reported above, and the determination of the spectral density function  $J_1(\omega)$  by Richardson et al.<sup>12</sup>. So far, theoretical calculations of  $J$  have been confined to the calculation of its energy. The energy calculations are in only fair agreement with experiment, especially in the BCC phase<sup>13</sup>. We would like to suggest that the results of calculations for the BCC

phase could be significantly improved if one took, in addition to the nearest neighbors, the next nearest neighbors into account.

In the BCC structure the distance of next nearest neighbors is only  $\frac{2}{\sqrt{3}}$  the distance of nearest neighbors and so contributes significantly to the energy even if one assumes a simple  $\frac{1}{r^6}$  interaction. This remark is prompted by the observation that the spectral density function  $J_1(\omega)$  for BCC solid  $\text{He}^3$  calculated by Richardson et al.<sup>12</sup> can be fitted by the sum of two Gaussians; one attributed to the nearest neighbors while the other is attributed to the next nearest neighbors. Figure 2 shows such a fit to the data of Figure 3 of reference 12. The solid line is given by the equation

$$J_1(\omega) \approx G_1^1(0) \exp\left[-\frac{1}{2} \left(\frac{\omega}{\omega_e1}\right)^2\right] + G_1^2(0) \exp\left[-\frac{1}{2} \left(\frac{\omega}{\omega_e2}\right)^2\right] \quad \text{or}$$

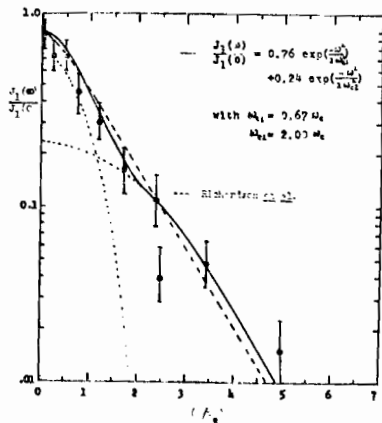


Figure 2

$$\frac{J_1(\omega)}{J_1(0)} \quad \text{vs} \quad \left(\frac{\omega}{\omega_e}\right)$$

$$10) \quad \frac{J_1(\omega)}{J_1(0)} = c \exp\left[-\frac{1}{2} \left(\frac{\omega}{\omega_e1}\right)^2\right] + d \exp\left[-\frac{1}{2} \left(\frac{\omega}{\omega_e2}\right)^2\right]$$

Since  $G_1^i(0) \propto \frac{z_i}{r_i^6}$  where the subscript 1 and 2 refer to the near-

est neighbors and next nearest neighbors, respectively,  $z_i$  is the number of neighbors and  $r_i$  is the interatomic distance

$$11) \frac{c}{d} = \frac{G_1^1(0)}{G_1^2(0)} = \frac{16}{5} \quad \text{for the BCC phase}$$

$$12) \quad c + d = 1, \text{ and we chose } \frac{\omega_{e2}}{\omega_{e1}} = 3$$

the dotted lines in Figure 2 represent the two Gaussians while the dashed line is the fit of Richardson et al<sup>12</sup> to the equation

$$13) \quad \frac{J_1(\omega)}{J_1(0)} = \exp \left[ \frac{-a(\omega)^2}{\omega_e^2 + b} \right] \quad \text{with} \quad \begin{array}{l} a = 1.023 \\ b = 0.178 \end{array}$$

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Discussion A10

I.A10 (L. H. Nosanow)

Q. P. M. Platzman (Bell Labs) Is there any way of estimating how important are the terms which are left out of the theory i.e. higher terms in cluster expansion and in exchange?

A. Yes. The 3-body cluster terms in the energy have been calculated. They are two orders of magnitude smaller than the 2-body term for correlation functions II and III. The correction terms for the exchange have not been calculated but are probably small.

Comment G. Meissner (Cornell) Instead of using the Heisenberg model for solid  $^3\text{He}$  a more microscopic approach is possible by calculating the spin-spin correlation function for the many-body Hamiltonian self-consistently.

A10.1 (J. M. Homer, M. G. Richards)

Q. L. Nosanow (Minnesota) Could you comment on your stated error in  $\theta$  of  $\pm 5\text{m}^\circ\text{K}$ ?

A. We get  $\pm 5\text{m}^\circ\text{K}$  by tilting our  $1/\chi$  against T plot so that the high temperature points would be all 0.5% too high and our low temperature points 0.5% too low. Also results from several runs always reproduce to within  $5\text{m}^\circ\text{K}$ .

Q. W. Fairbank (Stanford) Why didn't you go down to lower concentrations? We find a Curie law at 1 part in  $10^6$   $\text{He}^4$  in  $\text{He}^3$  and a small antiferromagnetic Weiss constant at 1 part in  $10^4$ . These are preliminary results and we are taking more measurements.

A. .06%  $\text{He}^4$  was the purest  $\text{He}^3$  we had available. We could go to lower concentrations but Nosanow says you would expect bulk J to increase proportional to concentration up to around 1%. Certainly, in the event of there being a large effect we would have expected to see the change as we increased  $\text{He}^4$  concentration.

Q. J. C. Wheatley (La Jolla) 1. How large was the quantity  $K \Delta V/V_0$  in the experiment? 2. How was the thermometry performed? 3. In fact the area under the resonance line did vary substantially from a  $1/T$  dependence.

A. 1.  $K \Delta V/V_0 = 1/4 \times 0.25 \approx 0.06$ . 2.  $\text{He}^3$  vapour pressure measurements with Macleod gauge. 3. Yes. In fact you would probably get quite a good straight line for  $K = 0$  but the value of  $\theta$  is very dependent on J.

Q. D. O. Edwards (Ohio) Did you observe anything interesting in the 45% mixture at the lowest temperatures? In the specific heat there is a pronounced 'tail' well above the

phase-separation temperature and this might show up in the susceptibility or relaxation time.

- A. We did not take measurements much below  $0.4^\circ\text{K}$  for the 45% mixture (i.e. not below the phase separation temperature). We found no large J to which the tail on the specific heat measurements might be attributed.

A10.2 (W. Senghaphan, G. O. Zimmerman)

- Q. M. G. Richards (Sussex) 1. What was the  $\text{He}^4$  content in this experiment? As little as 0.01% could affect some of the results. 2. What experimental check do you have that the sample as a whole was not being heated by the ultrasonic pulses? This could affect results both by heating the spin system and by shortening some of the relaxation times.
- A. 1. The  $\text{He}^3$  was purer than 99.9%, the next number being in doubt. We did not check our purity further because the main purpose of this experiment was to investigate the interaction of sound with the spin system which we felt would not be influenced by  $\text{He}^4$ . 2. We did observe some heating; however the heating could not account for our results.
- Q. R. C. Richardson (Cornell) What was the temperature of the measurement of the resonant coupling and what was the Larmor frequency?
- A. The temperature was between  $0.06$  and  $0.18^\circ\text{K}$ . The Larmor frequency was mainly 8.85 Mc/sec although near  $J/2\pi$  several frequencies were used and no frequency dependence observed within our scatter.

Comment H. Meyer (Duke) I question the validity of trying to fit the spectral density  $J_1(\omega)$  in BCC  $\text{He}^3$  by two gaussians due to nearest neighbours and next nearest neighbours respectively. The reasons are that the exchange between next nearest neighbours can be expected to be much smaller than between nearest neighbours, because in such an exchange,  $\text{He}^3$  atoms have to squeeze by 4 nearest neighbours. Hence the range of exchange cannot be estimated in terms of the ratio  $2/\sqrt{3}$  of the distances. I also feel that the form of  $G'$  is not correct. The shape of  $J_1(\omega)$  can be expected to depend on the number of nearest neighbours. For the closest packing of  $Z = 12$  nearest neighbours,  $J_1(\omega)$  can be thought to be closer to a gaussian form than for BCC (8 nearest neighbours). It would be most interesting to calculate this function for both lattices and also for simple cubic and diamond structures to study the influence of  $Z$  on  $J_1(\omega)$  for a Heisenberg type exchange.