Thermodynamic measurements of the multiple-exchange frequencies in bcc ³He

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We have studied the multiple-exchange Hamiltonian describing solid ³He by making a detailed comparison between the experiment and the predictions of the multiple exchange model at high magnetic field ($0 \le H \le 7.5$ T). The characteristic exchange frequencies are obtained by fitting the experimental data with the results of a statistical calculation using a high-temperature series expansion of the Hamiltonian in a temperature range where the magnetic effect is significant and the expansion converges rapidly.

I. INTRODUCTION

The simple description of the atomic exchange in solid ³He by a two-particle Hamiltonian has long since proved to be inadequate. This model, which gives a satisfactory description of the thermodynamic and relaxation properties of the solid at temperatures much larger than the antiferromagnetic transition, fails whenever the exchange dominates the properties of the solid, i.e., at temperatures near and below the transition or at high magnetic field when the spin polarization becomes significant.[1]

Several mechanisms have been proposed to explain the low-temperature experiments but most of these could only account for part of the data. The only model giving a complete and coherent picture of all the new results is a description of the atomic exchange including not only the exchange of two neighboring atoms but also the cyclic exchange of three, four, and possibly more atoms. However, a detailed comparison of the predictions of the model with the experimental results was still needed in a temperature range where theory and experiment could be reliably compared. This condition is fulfilled for pressure measurements at various magnetic fields in a proper temperature range. (1) At low temperature ($5T_N < T < 30T_N$, where T_N is the ordering temperature of the solid) the contribution of the phonons and vacancies to the pressure is negligible and the only significant contribution is due to the nuclear spins. Due to the nature of the atomic exchange in this fermion system this contribution is a strong function of the spin polarization $\bar{p}$ which can be varied over a wide range by applying a magnetic field ($0 \le H \le 7.5$ T). (2) Using statistical thermodynamics the pressure is deduced from a high-temperature series expansion of the partition function of the system and the temperature range is well enough above T_N to ensure that the expansion converges rapidly. Two earlier experiments have been carried out in a magnetic field[2,3] in a higher temperature range ($T > 20$ mK) where the effect is smaller. They were analyzed in the framework of the Heisenberg model[2] and a two parameter (J_t, K_p) multiple exchange model.[3]

A multiple-exchange Hamiltonian can be written quite generally

$$\mathcal{H}_{ex} = \sum_p (-1)^p J_p P^s , \qquad (1)$$

where the sum is extended over all permutations acting on spin variables, p is the permutation parity, and J_p are the characteristic exchange frequencies which, thus defined, are negative. In bcc ³He, the dominant processes are pair exchanges between first (J_{NN}) and second (J_{NNN}) neighbors, triple exchange J_t, and planar four-spin exchange K_p. Expressing the permutation operator of three spin-$\frac{1}{2}$ particles as a sum of two particle operators, the Hamiltonian (1) can be written as

$$\mathcal{H}_{ex} = -\tilde{J}_1 \overset{(1)}{\underset{\substack{i,j \\ i<j}}{\sum}} P_{ij} - \tilde{J}_2 \overset{(2)}{\underset{\substack{i,j \\ i<j}}{\sum}} P_{ij}$$
$$- K_p \underset{\substack{i,j,k,l \\ i<j<k<l}}{\sum} (P_{ijkl} + P_{ij\bar{kl}}) + \text{const} ,$$

where the effective pair exchange frequencies $\tilde{J}_1$ and $\tilde{J}_2$ are given by $\tilde{J}_1 = J_{NN} - 6J_t$, $\tilde{J}_2 = J_{NNN} - 4J_t$. The sums (1) and (2) are performed over first and second neighbors, respectively.

In our temperature range, the main energy reservoirs of the solid are the Zeeman and exchange systems whose Hamiltonians commute with one another. For an arbitrary magnetic field H, the partition function is given by $Z = \text{tr}[\exp(-\beta\mathcal{H}_{ex})\exp(-\beta\mathcal{H}_z)]$ with $\mathcal{H}_z = -(\gamma\hbar/2) \times H\sum_i \sigma_i^z$, $\beta = 1/k_B T$ where σ_i are the Pauli spin operators.

Expanding $\exp(-\beta\mathcal{H}_{ex})$ in powers of β and taking the logarithm, we obtain the contribution of the exchange to the pressure in any magnetic field through the volume derivative $P = k_B T \, \partial\ln Z/\partial V$ with

$$N^{-1}\ln Z = N^{-1}\ln Z_0 - e_1(\bar{p}^2)\beta$$
$$+ (1-\bar{p}^2)\sum_{n=2}^{\infty} \frac{a_n(\bar{p}^2)}{n!}\left[\frac{\beta}{2}\right]^n ,$$

and

$$e_1(\bar{p}^2) = -\left[(4\tilde{J}_1 + 3\tilde{J}_2 + 18K_p)\frac{\bar{p}^2}{2} + \tfrac{3}{2}K_p\bar{p}^4\right] ,$$

$$Z_0 = \text{tr}\exp(-\beta\mathcal{H}_z) = \left[2\cosh\left[\beta\frac{\gamma\hbar H}{2}\right]\right]^N .$$

The coefficients $a_n(\bar{p}^2)$ of the high-temperature expan-

sion of ln$Z$ are given up to the fourth order in Ref. 4. Note that in $e_1(\bar{p}^2)$ we neglect a physically unimportant constant term arising from the choice of the origin of the energies in the exchange Hamiltonian.

The best set of parameters $\bar{J}_1$, $\bar{J}_2$, K_p corresponding to a given sample is then obtained by fitting the theoretical value of P to the experimental data by minimization methods on a computer. We made the assumption that the molar volume dependences of the exchange parameters were similar and could be taken as V^{18}. Such dependences are expected both from theoretical considerations[5] and from measurements of various combinations of the parameters.[6-8]

II. APPARATUS

The cell is made of Stycast 1266 and placed in the Stycast mixing chamber of a dilution refrigerator. The thermal contact with the mixing chamber is made on the top and at the bottom of the sample space by sintered copper thermally connected to a sintered copper exchanger on top of the cell. The pressure is measured by a capacitive beryllium copper strain gauge located in the middle of the cell and surrounded by the solid. Keeping the dilution refrigerator at 4.2 K in order to leave the filling capillary of the cell open, the gauge is calibrated in liquid ^{3}He against two gauges at room temperature: a Sedeme CMB 200 gauge and a Digiquartz 2900AS gauge. The readings of the Be-Cu strain gauge have also been checked to be independent of the magnetic field.

Three sintered copper cylinders, thermally isolated from the sinter used for cooling the sample and placed in the upper part of the cell, are connected to the plates and shield of a SiO$_2$ capacitive thermometer located in a sealed enclosure on top of the cell. The measurement of the dielectric constant of the SiO$_2$ as a function of temperature is used as a secondary thermometer which is field independent.[9] One of the sintered copper cylinders, connected to the shield of the SiO$_2$ thermometer is thermally connected to a copper wire bundle immersed in the mixing chamber liquid on top of the cell. Continuous-wave NMR performed on this bundle is also used as a secondary thermometer. Both thermometers are calibrated by measuring the temperatures of melting curve ^{3}He samples and using the data of Ref. 10.

Two capacitive bridges are used for the measurements, one is a GR 1615A and the other, homemade, is based on the same principle as the GR bridge. Both bridges are used in connection with lock-in amplifiers.

The solid ^{3}He sample is prepared by a "blocked capillary" method, its temperature is raised close to the melting curve, the sample is then melted and solidified a few times before being annealed close to the melting curve for more than 10 h, and cooled slowly to the temperature range of the measurements. These samples are mainly bulk samples and typical cooling lengths are of the order of 1 mm. Taking into account the diffusion constants and the typical relaxation mechanisms in the solid,[11] the equilibrium times for our large molar volume bcc samples are at most of the order of one day. We typically waited three days before taking data after a temperature change.

The ^{4}He concentration of our samples is 60 ppm corre-

sponding to a phase separation temperature of the order of 60 mK (Ref. 11) which is well above the temperature range of our experiments. Any small pressure change due to the phase separation appears only once during the cooling down and does not interfere with our measurements.

III. RESULTS AND DISCUSSION

We carried our measurements of the pressure of two samples of molar volumes $V = 24.085$ and 23.610 cm^3 as a function of temperature in four applied magnetic fields, 0, 4, 6, and 7.5 T. The experimental data are plotted in Figs.
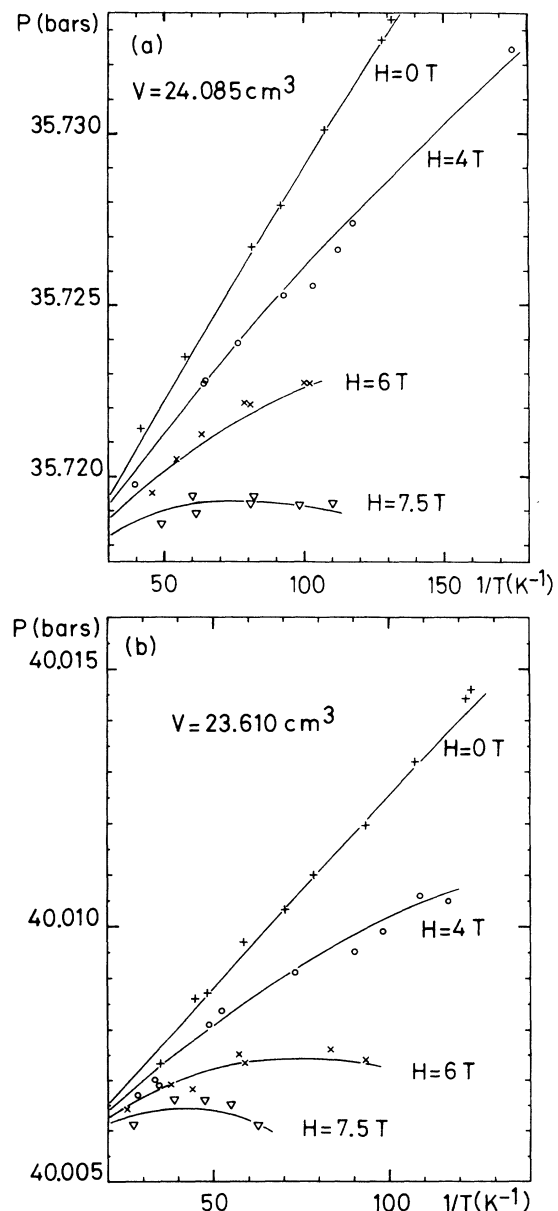

FIG. 1. Pressure in bcc ^{3}He samples of molar volume V: (a) $V = 24.085$ cm^3, (b) $V = 23.610$ cm^3, as a function of temperature at four magnetic fields. The solid curves are the fits made with the parameters of Table I.

1(a) and 1(b) together with the theoretical fits obtained by a method of least squares on a computer. As a first step we look for the best fit at constant K_p and then vary K_p continuously. We plot on Fig. 2 the set of parameters $\tilde{J}_1$ and $\tilde{J}_2$ giving the smallest rms deviation as a function of K_p together with the values of the rms deviation and the corresponding value of the Curie-Weiss temperature Θ for $V = 24.085$ cm^3. We obtain curves similar in shape for $V = 23.610$ cm^3. It appears that (1) the rms deviation goes through a smooth minimum as a function of K_p in the range $-0.45 < K_p < -0.3$ mK at $V = 24.085$ cm^3 and $-0.33 < K_p < -0.13$ mK at $V = 23.610$ cm^3; (2) $\tilde{J}_2$ is roughly constant for the whole range of K_p; and (3) θ is also fairly constant for the same range of K_p.

We thus get well defined values of $\tilde{J}_2$. The minimum of the rms deviation as a function of K_p being smooth, we obtain a small range of couples $(\tilde{J}_1, K_p)$ which could fit the data within the experimental uncertainties.

To further restrict the range of possible pairs $(\tilde{J}_1, K_p)$, we look for a physical parameter sensitive to a small change of $(\tilde{J}_1, K_p)$. One possible choice is the zero-temperature critical field H_{c1} between the two ordered phases. A mean-field calculation is not expected to give a very reliable result; however, the order of magnitude remains correct. Better estimates of H_{c1} are obtained when introducing the spin-wave approximation which gives larger values than those calculated with the mean-field approximation. We thus choose a theoretical mean-field value of H_{c1} in the range $0 \leq H_{c1} \leq 0.4$ T. The variation of H_{c1} with small changes in the parameters $(\tilde{J}_1, K_p)$ being large, i.e., when K_p varies from -0.35 to -0.38 mK,
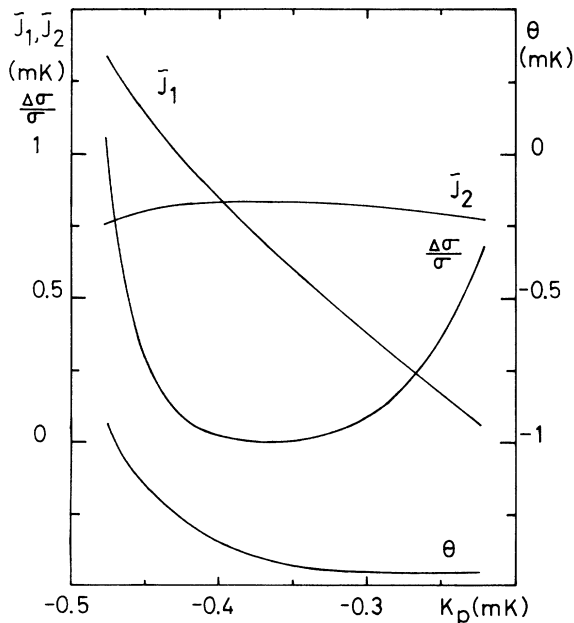
H_{c1} varies from 0.06 to 0.4 T and the possible values of $(\tilde{J}_1, K_p)$ are much restricted. It is worth pointing out that the values thus obtained and given in Table I agree with the minimum of the rms deviation. Taking into account the expected hierarchy of the parameters,[5,16] we choose $J_{NNN} = 0$ and obtain J_{NN}, J_t, and K_p.

Our values of e_2 are in good agreement with the measurements of Refs. 7, 12, and 13 and larger than the values of Refs. 3, 6, and 14. The figures of the Curie-Weiss constant Θ are also in good agreement with the most recent measurements of Refs. 3 and 15. Our ratios $J_t/J_{NN} \sim 0.35$ and $K_p/J_{NN} \sim 0.61$ are similar to those obtained by path-integral Monte Carlo calculations[16] while our exchange frequencies are 30% larger than the theoretical ones. Our values are also somewhat larger than the one deduced in Ref. 17 from a fit of the measured magnetic properties of the phase diagram of solid ^{3}He made in the framework of the mean-field theory adjusting e_2 to the value 6.8 mK^2, i.e., 35% lower than ours.

Using the parameters of Table I we have calculated several physical quantities and compared the results to the experiment. The figures are also given in Table I for our two molar volumes. The agreement between the calculated and measured properties is good.

We have only investigated two molar volumes and it is difficult to deduce significant Grüneisen-type constants for the variations of the measured parameters. The figures $\gamma_\Theta = 18.2$, $\gamma_{e_2} = 31.9$ are only estimates which are consistent with our initial assumptions.

As mentioned above, we have chosen to restrict our

FIG. 2. Two- and three-particle exchange frequencies $\tilde{J}_1$ and $\tilde{J}_2$, Curie-Weiss temperature Θ, and relative amplitude of the rms deviation $\Delta\sigma/\sigma$ corresponding to the best fit of our data at $V = 24.085$ cm^3 as functions of the four-particle exchange frequency K_p.

TABLE I. This table gives our measurements of the exchange frequencies for two molar volumes and some physical quantities which can be deduced theoretically. The notations used are standard and can be found in Ref. 1. The figures followed by an asterisk are values calculated for $V = 24.2$ cm^3 using $\gamma_{\tilde{J}_1, \tilde{J}_2, K_p} = 18$.

V (cm^3/mol)	23.610	24.085
$\tilde{J}_1$ (mK)	0.485	0.664
$\tilde{J}_2$ (mK)	0.614	0.833
K_p (mK)	-0.264	-0.364
J_{NN} (mK)	-0.435	-0.585
J_t (mK)	-0.153	-0.208
Θ (mK)	-0.971	-1.396
T_{c1} (mK)	1.014	1.388
T_{c2} (mK)	1.485	2.024
C_1/χ (mK)	3.170	4.413
	4.943*	4.808*
B (mK^2)	0.035	0.022
e_2 (mK^2)	4.740	8.940
e_3 (mK^3)	-1.836	-4.872
H_{c1} (T)	0.138	0.214
H_{c2} (T)	11.304	15.623
	17.627*	17.022*
C_{SW} (cm/sec)	6.261	8.759
	9.764*	9.543*

analysis of the data to three parameters which are expected to be the most significant on theoretical grounds.[5,16] The fit of the data with the theory is good and we never needed to introduce higher-order exchanges. This same set of three exchange frequencies also accounts quantitatively for most of the known properties of the low-temperature phases as shown in Table I. We must keep in mind that some of these quantities are very difficult to calculate theoretically and can only be estimated.

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