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Specific heat measurements on bcc ^3He from 3 to 300 mK

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Résumé. — Nous avons mesuré la chaleur spécifique de ^3He solide ($23,8 \text{ cm}^3/\text{mole}$) de 3 à 300 mK. On obtient une valeur constante de l'intégrale d'échange, égale à 0,56 mK, en très bon accord avec les résultats à haute température obtenus sur un échantillon massif. Aucune contribution notable liée à la présence de polarons magnétiques n'apparaît en dessous de 50 mK. Un excès de chaleur spécifique est par contre observé vers 100 mK.

Abstract. — We have measured the specific heat of bcc ^3He ($23.8 \text{ cm}^3/\text{mole}$) from 3 to 300 mK. A constant value, 0.56 mK, is obtained for the exchange integral, in very good agreement with higher temperature bulk results. No contribution due to magnetic polarons appears below 50 mK. An excess in the specific heat is observed around 100 mK.

It has recently been emphasized [1] that the specific heat [2] of bcc ^3He (below 20 mK) at $23.8 \text{ cm}^3/\text{mole}$ confined in sintered copper (grain size $\sim 1 \mu$) exhibit an excess with respect to the magnetic exchange term deduced from experiments on bulk ^3He above 40 mK [3]. An indirect determination of the specific heat of ^3He seems to confirm this result [4]. A vacancy magnetic polarons model leading to a very large anomaly in specific heat between 20 and 40 mK has been developed and accounts for these results using a large concentration of vacancies : $\sim 7 \times 10^{-4}$ [1].

We have measured the specific heat of bcc ^3He ($< 50 \text{ ppm } ^4\text{He}$) at $23.8 \pm 0.1 \text{ cm}^3/\text{mole}$, in the range 3-300 mK, in a demagnetization cell of CMN attached through a superconducting heat switch to a dilution refrigerator [5]. An araldite cell

$$(\varnothing 28.5 \text{ mm}, h = 28.5 \text{ mm})$$

designed for pressures larger than 50 bars contains 21.85 g of CMN (grain size $< 40 \mu$) packed at a filling factor of 67 %. An Evanohm heater ($\varnothing 50 \mu$, about 90 cm long) is distributed in the CMN in order to have no point in the cell further than about 2 mm from the heater. Its resistance, measured at 4 K, 100 mK and 30 mK, is $505.5 \pm 0.2 \Omega$. The temperature of the cell is measured above 15 mK through a 100Ω Speer sliced carbon resistor and below 40 mK using the susceptibility of the cooling CMN calibrated against the carbon resistor. This susceptibility follows a Curie law down to about 5 mK [6]. As previously

measured [5, 7] in such experimental arrangements, the magnetic temperature indicated by the CMN is about 0.5 mK less than the thermodynamic temperature at the transition of superfluid ^3He near the melting pressure.

The cell was filled with liquid ^3He at a pressure of 48 bars at 1.4 K. The 1 kG magnetizing field was then applied and the precooling process started. The solid was grown in about 2 hours and reached 100 mK 12 hours later. After two days precooling the demagnetization of the CMN was performed starting from about 16 mK. The final temperature was around 2.5 mK and the heat leak about 2 ergs/min. Under those conditions the specific heat data were obtained using the classical adiabatic procedure. The total heat capacity is deduced from the ratio of the supplied energy ΔQ to the corresponding increase ΔT of the temperature. The ratio $\Delta T/T$ was varied from 1 to 10 % and for a given energy ΔQ we varied the power by a factor of 100, without obtaining any noticeable difference in the specific heat results.

In the analysis of the data, below 12 mK, the background heat capacity, essentially the CMN one, is that deduced from experiments with liquid ^3He or a ^3He - ^4He mixture as a thermal contact agent [6, 8]. In the range 12 mK-70 mK, the background was measured by filling the cell with liquid ^4He at 1 bar. The results are in very good agreement with previous data [5, 6, 9, 10]. Above 70 mK the heat leak through the filling capillary was too large due to the very high ^4He thermal conductivity and other CMN data [10] have been used, estimating that the heat capacity of

the araldite cell and the addenda remains negligible. This is confirmed by the measurement of the bcc ^3He Debye temperature as may be seen below.

In order to know the volume of the cell, we performed also an experiment with liquid ^3He at 29.4 bars in the range 4-200 mK. Scaling with liquid ^3He values at very low temperatures [11], we deduced the number of moles : 0.295 ± 0.01 corresponding to a volume of the cell equal to $7.70 \pm 0.25 \text{ cm}^3$. In view of the filling factor of the cell with the CMN, a dead volume of bulk He, at most $\sim 0.5 \text{ mm}$ thick, is probably located at the bottom of the cell.

The specific heat results are presented on figure 1 as a function of the temperature. Below 50 mK, down to the lowest temperature, the specific heat varies as T^{-2} as may be seen on figure 2 where we have plotted CT^2 versus T^{-1} . This is expected from a simple

Fig. 1. — Specific heat of bcc ^3He ($23.8 \text{ cm}^3/\text{mole}$) versus temperature. The solid curve represents the sum of the exchange and the Debye terms.

Fig. 2. — $CT^2(T^{-1})$ plot for bcc ^3He . The dashed line represents the pair exchange term. The solid curve corresponds to the least square fitted results of ref. [2] : due to the large error limits reported by the authors (shaded area), the difference with our results is probably less significant than the curves seem to show.

Heisenberg nearest neighbour (HNN) model assuming that the exchange arises from the nearest neighbour pairs with an exchange energy J . The specific heat is then given by :

$$C_v = R[3(J/k_B T)^2 + \dots]$$

Our results yields to : $|J|/k_B = 0.56 \pm 0.01 \text{ mK}$. As seen on figure 3, this value is in very good agreement with previous results [3, 12-16].

Fig. 3. — Exchange integral versus molar volume (from Ref. [3]).
 ▼ This work ; ▽ ref. [12] ; ● ref. [3] ; □ ref. [13] ; ◇ ref. [14] ;
 ○ ref. [15] ; △ ref. [16].

At the lowest temperatures, when one approaches magnetic ordering ($T_N \simeq 1 \text{ mK}$), the influence of higher order terms in the expansion of the specific heat has to be considered [17]. Down to 4 mK no significant departure from a T^{-2} variation appears. Below 4 mK the accuracy of our measurements decreases due to the lack of thermal contact and to the uncertainty on the temperature scale which is no longer the thermodynamic one. However a small deviation seems to appear at the lowest temperature, giving a negative sign to the third order term in agreement with previous results [2, 4], confirming the inadequacy of the HNN model to describe the millikelvin properties of bcc ^3He . Using the notation of the four spin exchange model [18, 19] :

$$C_v = \frac{R}{4} \left(\frac{\tilde{e}_2}{T^2} - \frac{\tilde{e}_3}{T^3} \right)$$

we obtain : $\tilde{e}_2 = 3.8 (\text{mK})^2$ and a *maximum* value for $\tilde{e}_3 \lesssim 2.0 (\text{mK})^3$. This agrees with the idea that the four spin terms are at most of the same order of magnitude as the pair exchange term [20]. This is also in agreement with a non-linear lattice-dynamical

approach [21] where no large entropy effects are expected at relatively large temperature compared to the ordering temperature. Nevertheless the *maximum* value of the third moment $J_{xxx}^3 (= 1/12 \tilde{\epsilon}_3)$ of the expansion, which we determine as 0.17 (mK)^3 , is five times smaller than that obtained in other experiments [2].

In the range 10-50 mK we do not observe any anomaly in the specific heat as seen in figure 2 where our results are compared with previous experiments [2]. No evidence is found for a large contribution of magnetic polarons to the specific heat in this temperature range [1, 22]. In order to take place within error bars, the concentration of vacancies calculated from the model would have to be less than 5 % of that proposed [1] to explain the results obtained in a smaller copper powder [2].

We should point out that the size of the helium channels is about 20 times larger in our cell. Either the fact that these conditions are nearer those of the bulk or other experimental features (cooling process for example) may explain the difference in the results.

From the recovery time after small demagnetizations [5], we have also studied the heat transfer between the CMN and the solid below 30 mK. Using the classical two bath model, a constant Kapitza resistance is obtained below 20 mK :

$$R_K \simeq 0.35 \text{ s.mK/erg},$$

which is in qualitative agreement with previous heat transfer measurements between liquid ^3He and CMN with the same grain size [12], as well as solid ^3He -CMN (larger grain size) experiments [23]. This confirms an anomalously small contact resistance between ^3He and CMN, but the absence of a temperature dependence does not correspond to the *magnetic coupling* calculations where a resistance proportional to T^2 is calculated, both susceptibilities of solid ^3He and CMN varying as T^{-1} in this temperature range [24, 25].

Turning now to the highest temperatures we observe a specific heat varying as T^3 , as expected for a Debye regime with a characteristic temperature :

$$\theta_D = 18.0 \pm 0.5 \text{ K} \quad \text{at} \quad \frac{T}{\theta_D} \simeq 0.011.$$

This value is in good agreement with other measurements on bcc ^3He performed in a cell where the size of the channels ranged around 100μ [13], but lies 10 % smaller than that obtained in bulk solid [3]. This is related to the presence of an excess specific heat in the sum of the exchange and the Debye terms seen on figure 1. At 120 mK this difference is about 40 % of the measured value, roughly the same as previously reported [13]. Nevertheless the anomaly here does not fit a T term but appears nearly constant between 80 and 200 mK [26]. The large increase of CT^2 for $T > 50 \text{ mK}$ (Fig. 2) corresponds to this anomaly. This contribution to the specific heat around 100 mK is observed when the sample dimensions are in the submillimeter range. The physical origin of such an anomaly may be possibly related to the presence of defects in these types of geometry : zero point vacancies, randomly dispersed vacancies, etc. Varying different parameters (cooling process, size of the grains, packing factor) should give more precise data for analysing this problem. Experiments are in progress to get more information on this *high temperature anomaly*.

As a conclusion, it follows from these specific heat experiments, that bcc ^3He appears as a good paramagnet down to 4-5 mK without evidence of an approach to ordering, in agreement with susceptibility measurements [27, 28] which follow a Curie-Weiss law in the same temperature range. No large contribution of magnetic polarons has been observed in these experiments but an excess in specific heat is measured around 100 mK in this confined geometry.

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