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SELF DIFFUSION IN SOLID ^3He AT VERY LOW TEMPERATURE

G. DEVILLE, J. M. DELRIEU and M. BERNIER

DPh-G/PSRM, CEN - Saclay, B.P. 2, 91190 Gif sur Yvette, France

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Résumé. — Les échos multiples dans l' ^3He solide, dus à l'effet du champ démagnétisant, ont été recalculés en tenant compte de la diffusion de spin et nous avons appliqué cette analyse à des résultats expérimentaux antérieurs obtenus à très basse température. Le coefficient de diffusion de spin D reste constant dans tout le domaine de température 10,5 mK-1,07 mK, même au voisinage de la température d'ordre des spins de l' ^3He solide. On détermine les valeurs de la susceptibilité magnétique χ avec une précision améliorée par rapport aux valeurs précédemment publiées [3].

Abstract. — Multiple spin echoes in solid ^3He , which are due to the demagnetizing field effect, have been reconsidered by taking into account the spin diffusion. The analysis is applied to the experimental results obtained earlier at very low temperature. The spin diffusion coefficient D remains constant in the whole temperature range 10.5 mK-1.07 mK, even close to the spin ordering temperature of solid ^3He . The susceptibility values χ obtained have a higher precision than those previously published [3].

From an earlier analysis [1, 2, 3] of N.M.R. *multiple echoes* obtained at low frequency in solid ^3He prepared in a Pomeranchuk cell in a temperature range 1-20 mK, we deduced values of the longitudinal and transverse relaxation times T_1 and T_2 together with an absolute measurement of the susceptibility χ as a function of the temperature T . The analysis neglected spin diffusion in the solid. In this letter we include the effect of diffusion and this more refined analysis gives the value of the diffusion coefficient D together with more consistent figures for T_1 , T_2 and χ although they are only slightly different from those of the earlier analysis.

We recall the physical principles of the *multiple echoes* before discussing the new analysis of the experimental results.

1. Principles of the « multiple echoes ». — Multiple echoes appear as a response of the spin system of solid ^3He to a sequence of two radio-frequency pulses of any angle produced at times 0 and τ . The spin Hamiltonian of the system is the sum of Zeeman, exchange and dipolar contributions. The long range effect of the dipolar Hamiltonian, equivalent to the demagnetizing field H_d , is usually negligible in N.M.R. and consequently the above sequence is followed by just one echo. In solid ^3He , due to the exchange narrowing of the line, T_2 is long and the effect of

H_d becomes significant since $\gamma H_d T_2 \geq 1$. The evolution of the local magnetization $\vec{M}(\vec{r})$ is given by [4]

$$\frac{d\vec{M}(\vec{r})}{dt} = \gamma \vec{M}(\vec{r}) \wedge [\vec{H}(\vec{r}) + \vec{H}_d(\vec{r})] - \frac{M_z - M_0}{T_1} \vec{z} - \frac{M_x \vec{x} + M_y \vec{y}}{T_2} + D \nabla^2 \vec{M}(\vec{r}) \quad (1)$$

where $H(\vec{r})$ is the applied magnetic field.

$\vec{H}_d(\vec{r})$ is usually a tensorial function of $\vec{M}(\vec{r})$. This makes the equation (1) non linear and the mixing of different spatial Fourier components produces multiple echoes at times $n\tau$ (n being an integer, $n \geq 2$). When using a uniform magnetic field gradient G along the direction $\hat{s}$ superimposed on the applied field $\vec{H}_0$, $\vec{H}_d(\vec{r})$ becomes a linear function of $\vec{M}(\vec{r})$ [2, 5]

$$\vec{H}_d(\hat{s}, \vec{r}) = \beta M_z(\hat{s}, \vec{r}) \hat{z} + \frac{\beta}{3} \vec{M}(\hat{s}, \vec{r})$$

where $\beta = 2\pi[3(\hat{s} \cdot \hat{z})^2 - 1]$.

In this case an analytic solution of equation (1) is obtained when $D = 0$ [2]. When taking into account the diffusion, we calculate the time evolution of the spatial Fourier transform components of the magnetization using a computer HP 9025A [5] and we

obtain the amplitudes of the successive echoes. The values of T_2 , χ and D are the adjustable parameters obtained upon fitting the experimental data to the theory.

2. Experimental results. — First, let us recall the main experimental conditions. The sample of solid ^3He was formed in a Pomeranchuk cell and consequently was in contact with liquid ^3He along the melting curve ($V_m \approx 24.25 \text{ cm}^3$). The N.M.R. spectrometer used a pair of crossed coils, both the main magnetic field H_0 and the gradient were produced by the same Helmholtz pair. Owing to the large dimensions of these coils, both H_0 and G were quite uniform on the whole sample, G ranging from 0.2 G cm^{-1} to 2 G cm^{-1} and H_0 from 19 G to 79 G .

FIG. 1a. — Experimental amplitudes of multiple echoes following a two 90° pulses sequence, obtained in solid ^3He ($V_m = 24.25 \text{ cm}^3$) at 1.47 mK . The external field gradient was 0.8 G cm^{-1} . The time scale is in units of τ/T_2 where τ is the time interval between the two pulses and T_2 is deduced from the fit ($T_2 = 250 \text{ ms}$). The amplitudes A are normalized with the free precession amplitude A_0 following a 90° pulse after complete recovery. We can observe more than 30 echoes when $\tau/T_2 \approx 0.2$ but we have only reported 4 echo amplitudes for the sake of clarity. The lines through the experimental points are an aid for the eye.

Figure 1 gives an example of the comparison between theory and experiment. In figure 1a we plot the amplitudes of some of the multiples echoes as a function of τ . The theoretical amplitudes obtained using the theory including the diffusion are shown in figure 1b. Figure 1c illustrates the comparison between the experimental amplitudes of echo (1) and those calculated both with and without diffusion. Taking the diffusion into account significantly improves the agreement between theory and experiment at large values of τ . The fit was made by adjusting the 3 parameters T_2 , χ and D at each temperature. We point out that T_2 thus obtained is equal to T_1 within the experimental errors ($T_1 = T_2 = 250 \text{ ms}$), in agreement with high temperature results [6] and does not vary with temperature. The values of $1/\chi$ and D are plotted on figure 2. The straight line is drawn with the Curie-Weiss constant $\theta = 2.6 \text{ mK}$. This Curie-Weiss temperature enables us to compare our results

FIG. 1b. — Calculated echo amplitudes with the theory including the diffusion effect. The adjusted parameters T_2 , χ and D are $T_2 = 250 \text{ ms}$, $\chi = 0.81 \times 10^{-5}$ and $D = 15 \times 10^{-8} \text{ cm}^2 \cdot \text{s}^{-1}$. $T_1 = 250 \text{ ms}$ was measured independently.

FIG. 1c. — Comparison between experiment and theories with and without diffusion. This comparison is made with only one echo for the sake of clarity. The curve (1 α) gives the experimental amplitudes (see Fig. 1a). The curve (1 β) gives the calculated amplitudes when including the diffusion (see Fig. 1b). The curve (1 γ) gives the calculated amplitudes when neglecting the diffusion as they appear in figure 2 of reference [2].

FIG. 2. — Plot of the inverse susceptibility and of the diffusion coefficient as a function of the temperature. The Curie-Weiss law with $\theta = 2.6 \text{ mK}$ and the results of reference [7] are given for comparison. The error bars for D give the relative uncertainty of the measurement. The error on the absolute value of D is due to the lack of precise knowledge of the field gradient G as the N.M.R. experiments only give the product DG^2 .

with the Prewitt-Goodkind data [7] although these are obtained with a solid of slightly different molar volume. Nevertheless, as the multiple echoes method gives absolute values of χ , we are not obliged to normalize our low temperature data with the extrapolated high temperature data. Consequently, our $1/\chi$ results are plotted with an absolute scale while the Prewitt-Goodkind data are scaled according to the absolute Curie-Weiss law with $\theta = 2.6$ mK. The temperature scale in both experiments is deduced from secondary thermometers : N.M.R. on copper in reference [7], and the pressure along the melting curve in our experiments. Fixed points of our temperature scale are obtained in every run when crossing the well known A and B transitions of liquid ^3He . The small difference in transition temperatures between the two experiments : 1.25 mK in reference [7] and 1.07 mK in our experiment might be explained by experimental uncertainties.

The constant D values are plotted on the right vertical scale. Due to the lack of precise knowledge of the experimental field gradient, we cannot report an accurate absolute value of D ; we give $D : (15 \pm 5) 10^{-8} \text{ cm}^2 \cdot \text{s}^{-1}$. However, we think that, in the temperature range 10.5 mK-1.07 mK, D is constant to better than 10 %, this corresponding to the error bars shown in figure 2. This new result is in contradiction with the sole earlier measurement of T_2

and D [8] made in the same temperature and frequency ranges. The apparent variation of T_2 and D reported in reference [8] is probably related to the existence of non reported multiple echoes which produce a fast decrease of the first echo as shown on figure 1a.

Let us point out that according to the time range the behaviour of the echoes is mainly dependent on one of the three main parameters χ , T_2 and D for short, intermediate and long times τ respectively [5]. The quality of the fit is not dependent on the temperature although in our experiment the magnetization varies by a factor of five and this gives us some confidence in our measurements.

If solid ^3He were a conventional Heisenberg antiferromagnet we could expect the diffusion to increase when approaching the transition from the paramagnetic phase as is calculated [9] and observed [10] in electronic antiferromagnetic spin systems. On the other hand, a decrease of D is expected in the qualitative dynamical approach of Papoular [11]. We think that the same remains probably true if there are spin polarons in ^3He [12]. It would be interesting to calculate the effects of four spin exchange [13] on D and T_2 : in this model the two opposite tendencies, ferromagnetic and antiferromagnetic, might produce some compensations. Another possible explanation is that the observed transition is first order without strong pretransitional effects on T_2 and D .

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