

The Exchange Narrowing of T_1 in Solid Helium-3

— The Fourth Moment of the Resonance Line —

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(Received December 3, 1987)

Abstract

The exchange narrowing of the nuclear magnetic relaxation time T_1 of solid ^3He is investigated on the basis of the heat bath model. The exchange interactions are supposed to be described by the ring exchange model with two or three parameters. It is found that both parameter models, the two and the three parameter models, can not explain the observed T_1 data. The Van Vleck's fourth moment of the hcp lattice is also obtained. This will be useful to establish a microscopic model of hcp ^3He .

§ 1. Introduction

The nuclear magnetism of solid ^3He has attracted much attention, because it exhibits more fascinating behavior than that previously thought. The most exciting feature is the existence of higher order spin interactions, such as four or six particle ones. Though the microscopic origin of such interactions is still

unclear, some theoretical models are proposed [1]. Among these models the ring exchange model, in which the exchange interactions are limited to the triple and the four particle ones, is very accepted. In fact, the most experimental data have been successfully explained by this ring exchange model [2]. Thermodynamic properties in the high temperature regime, such as the pressure, the specific heat, the magnetic susceptibility have been explained by this model. Furthermore, some aspects of magnetic ordered phases (LFP and HFP) are consistent with the predictions of the same model. On the other hand the multiple exchange models have not been applied to the theoretical analysis of the narrowing of NMR. In the early literature, the exchange narrowing was explained quite successfully on the basis of the Heisenberg model with the nearest neighbor interaction [3, 4]. However, the present understanding is quite different. The nearest neighbor exchange interaction is rather small while the four spin interaction is large. Then, there remains a question whether the ring exchange model with two or three parameters can really explain the exchange narrowing.

It should be noticed, here, that the four spin interaction is more effective on the narrowing than the pair spin interactions. Our concern is that the ring exchange model may give rise to the stronger narrowing than the experimentally observed one [5].

§ 2. The model Hamiltonian and the relaxation time

We follow here the widely accepted view that the four spin interactions are quite important in solid ^3He . To be more precise we take the Heisenberg model including the four spin interaction of the planar type,

$$\begin{aligned} H_{\text{ex}} = & -\sum_i \sum_j 2J_n(S_i \cdot S_j) - \sum_{ijkl} 4K_P S_{ijkl}, \\ S_{ijkl} = & (S_i \cdot S_j)(S_k \cdot S_l) + (S_i \cdot S_k)(S_j \cdot S_l) \\ & + \lambda(S_i \cdot S_l)(S_j \cdot S_k), \end{aligned}$$

where the exchange parameters J_1, J_2, J_3 are the effective couplings of the first, second, and third nearest neighbor pairs, respectively.

From the thermodynamical point of view the nuclear spin system of solid ^3He consists of mutually interacting subsystems. As such subsystems we mention the

system of phonons, that of vacancies, the Zeeman system, and the system of exchange tunnelings. The spin reversals caused by the high frequency field give rise to a temperature rise in the Zeeman system. The excess energy due to this temperature rise leaks into the other heat bath systems. In the sub-1K region, where T_1 is independent of temperatures, the Zeeman-exchange relaxation dominates dynamical properties of the nuclear spin system, i. e. it is a bottle neck of the energy flow from the Zeeman system to the final heat reservoir. In such a temperature region we can use the exchange narrowing theory based on the heat bath model. For powder sample, this heat bath model gives the relation between the longitudinal relaxation time T_1 and the spectral density function $J(\omega)$, i. e.

$$\frac{1}{T_1} = J(\omega) + 4J(2\omega),$$

where ω is the Larmore frequency. The spectral density function is the Fourier transform of the correlation function characterizing the exchange motion. The torques acting on the unclear spins under the Larmore precessions are assumed to be a Gaussian random process.

Then, the spectral density function is allowed to be

$$J(\omega) = \frac{\sqrt{2\pi}}{3} \frac{M_2}{\omega_e} \exp\left(-\frac{\omega^2}{2\omega_e^2}\right),$$

where the exchange frequency ω_e is defined as

$$\omega_e^2 = \frac{M_4}{M_2},$$

M_2 and M_4 are second and fourth moments of the resonance line. The second moment for bcc lattice is calculated in the early literature [3],

$$M_2 = (9/20) \times 29.04(\gamma^2 \hbar)^2 / d^6,$$

where d is the second nearest neighbor distance, and γ is the gyromagnetic ratio of ^3He nucleus.

The Zeeman system is coupled to the exchange system by the dipole-dipole interaction. This interaction Hd is decomposed into the sum $Hd = \sum Gm$, where Gm is the part of the dipolar interaction that increases the z-component of the

total spin by $m(=-2, \dots, 2)$. It is well known that the Van Vleck's fourth moment M_4 can be expressed as

$$M_4 = \langle \frac{\text{Tr}\{[\text{Hex}[Go, Ix]]^2\}}{\text{Tr}\{Ix^2\}} \rangle,$$

where the trace is taken over the spin variables and $\langle \rangle$ denotes the angular average.

§ 3. Results and Conclusion for bcc ^3He

The calculation of M_4 is carried out in terms of the diagrams. When the exchange bonds in a diagram do not entirely coincide, the dipole bonds leading to non-vanishing contribution are finite in number. For types of diagrams in which the exchange bonds coincide, an infinite number of the dipolar bonds can lead to a non-vanishing contribution. The contribution from this infinite series is important.

For any given spin in Ix , we have calculated the diagrams covering 2×24^3 lattice points around the given spin. Then up to 2×48^3 lattice points. (The correction is found to be $1.5 \times 10^{-2}\%$.) Finally we proceed up to 2×96^3 lattice points, finding the very small correction of $1.2 \times 10^{-3}\%$.

Thus we obtain the result,

$$\begin{aligned} M_4 = & 9 \frac{(\gamma^2 \hbar)^2}{d^6} \{ 33.2J_1^2 + 31.2J_2^2 + 72.5J_3^2 \\ & + 13.6J_1 \cdot J_2 + 13.5J_1 \cdot J_3 + 6.4J_2 \cdot J_3 \\ & + (176.7 + 113.3\lambda + 86.9\lambda^2)K_P^2 \\ & - [(15.2\lambda - 0.65)J_1 + (7.59 + 15.2\lambda)(J_2 + J_3)]K_P \}. \end{aligned}$$

In the early investigations, the experimental value of T_1 was explained by the nearest neighbor Heisenberg model with the parameter $J_{NN} = -0.75\text{mk}$ (at $24.2\text{cm}^3/\text{mol}$). This model gives $T_1(0) = 289\text{ msec}$, where $T_1(0)$ is the $\omega \rightarrow 0$ limit of the nuclear relaxation time T_1 . This value seems to be in agreement with that obtained in the early experiments.

In recent years, the successful model for the magnetism of solid ^3He was proposed, that is the ring exchange model. Then we have estimated $T_1(0)$ of this

model, noting that the effective spin couplings (J_1 , J_2 , and J_3) are related to the ring exchange parameters J_{NN} , J_t , K_P , through

$$J_1 = J_{NN} - 6J_t + 3K_P,$$

$$J_2 = -4J_t + K_P,$$

$$J_3 = K_P/2,$$

and

$$\lambda = -1.$$

For the two parameter model ($J_t = -0.13\text{mK}$, $K_P = -0.385\text{mK}$), we find $T_1(0) = 384$ msec. It seems that this value is rather larger than that of the nearest neighbor Heisenberg model.

There is another parameter set, i. e. the three parameter model of Stipdonk and Hetherington [6] with $J_{NN} = -0.377\text{mK}$, $J_t = -0.155\text{mK}$, and $K_P = -0.327\text{mK}$.

For this model we obtain $T_1(0) = 356$ msec. Therefore, it is concluded the two and three parameter models can not explain the observed relaxation time T_1 . This conclusion strongly suggests that it may be worthwhile to research other models or the exchange mechanism. Such investigations were carried out in some publications emphasizing on the importance of virtual phonon processes which induce the four or more higher order spin interactions.

§ 4. Comment on hcp ^3He

Solid ^3He undergoes a phase transition to the hcp phase at 106atm. There is a speculation that the magnetism of hcp ^3He is described by the simple nearest neighbor ferromagnetic Heisenberg model, because only the triple exchange processes are supposed to be dominant. However, this speculation does not seem to be confirmed at present. Then we believe that the possibility of the existence of more long ranged pair type interactions and four spin interactions has not been excluded. In this report, T_1 formula is obtained for the hcp lattice based on the more complicated model which includes the first and second nearest neighbor pair type interactions, and the square type of four spin interaction. This model is proposed by several authors to explain the observed susceptibility data which is well plotted by the Curie-Weiss law even in the quite low temperatures [7, 8].

The second moment of the resonance line for the hcp lattice is obtained as

$$M_2 = (9/20) \times 14.45(\gamma^2 \hbar)^2 / d^6.$$

Furthermore, the fourth moment is also obtained as follows,

$$\begin{aligned} M_4 = & 9 \frac{(\gamma^2 \hbar)^2}{d^6} \{ 14.9J_1^2 + 14.9J_1'^2 + 18.0J_2^2 \\ & + 0.59J_1J_1' - 0.55J_1J_2 + 6.78J_1J_2 \\ & + (77.5 + 45.6\lambda + 34.7\lambda^2)K_{sq}^2 \\ & + [(0.26 - 1.2\lambda)(J_1 + J_1') - (2.4 + 4.8\lambda)J_2]K_{sq} \}. \end{aligned}$$

Though we have no enough data to determine the exchange parameters (J_1 , J_1' , J_2 , J_3 , K_{sq} , λ) experimentally at present, many experiments and measurements on hcp ^3He will be carried out in the near future. Then this model and the results will be useful for the further investigation of hcp ^3He . Particularly, we hope that a novel experiment which reveals the more exotic nature of hcp ^3He , e. g. antiferromagnetic spin ordering, will be carried out.

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