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small θ , the component of field perpendicular to [110] is therefore approximately the same for both \vec{H}_i and \vec{H}_0 and is given by $H_0 \sin\theta \approx H_0\theta$, while the corresponding parallel components, though different, remain independent of θ to first order. The product $H_x H_y H_z$ is thus proportional to θ referred to either internal or external fields. In practice, it is clearly more convenient to use the angle of the applied field as a measure of the induced staggered field. A further discussion of this point is given in N. Giordano, thesis, Yale University, 1977 (unpublished).

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Theory of Interlayer Exchange for Adsorbed ^3He Monolayers

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(Received 23 June 1977)

An explanation of experimental NMR relaxation times, T_2 , for ^3He adsorbed on Grafoil at coverages just greater than one monolayer is given in terms of the dipolar interaction of first-layer particles modulated by quantum exchange of particles between first and second layers. It is found that a Heisenberg-Hamiltonian description fails and one must use an exchange operator formulation which permits transitions among second-layer \vec{k} states during the exchange process.

Helium adsorbed on Grafoil represents a quasi-two-dimensional system which has a number of interesting phases,¹ e.g., gas, liquid, solid, and registered solid. Recently, experimental NMR results² (Fig. 1) have shown evidence for three distinct phases at $T = 1$ K: (a) a fluid phase for coverage $x \lesssim 0.7$; (b) a two-dimensional solid for $0.7 \lesssim x \lesssim 1.0$, and (c) a phase for $x \gtrsim 1$ for which T_2 increases linearly with coverage. Theoretical calculations² for region (b) showed that the T_2 data could be explained in terms of a dipolar linewidth motionally narrowed by quantum tunneling. Region (c) of the T_2 data was tentatively identified as being caused by the formation of the second layer of helium atoms with motion via exchange with the first layer. Evidence for a registered

phase was also reported recently.³

In this Letter I report calculations which justify the explanation of region (c) in terms of interlayer exchange. The novel feature of this explanation is the discovery that a calculation based on a Heisenberg-exchange Hamiltonian proves inadequate and one needs an exchange operator formulation.^{4,5} The exchange operator is usually written in the form⁹

$$H_{\text{exch}} = \sum_P \mathcal{G}_P P^{(\sigma)}, \quad (1)$$

where $P^{(\sigma)}$ is a pair permutation operator in spin space and \mathcal{G}_P is an operator having diagonal and off-diagonal elements in position space (usually in a phonon basis). This concept was originally suggested by Thouless,⁴ and developed later by

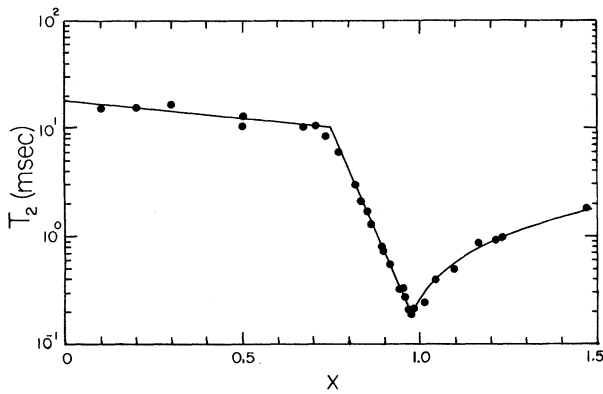


FIG. 1. Data from Ref. 2 showing values of T_2 vs coverage x . The region of interest for this paper is for $x \geq 1$. The data of this region are fitted quite well by the straight line $T_2 = 0.18 + 3.0(x - 0.98)$. The logarithmic scale distorts this linearity.

Nosanow and Varma,⁵ Guyer,⁶ McMahan,⁷ and Mullin⁸ for bulk ^3He systems. While the one-phonon matrix element of \mathcal{J}_P can be related to the magnetic Grüneisen constant⁶ and the two-phonon matrix element to spin-lattice relaxation,⁵ it is probably safe to say that this concept has not occupied a particularly vital place in the theory of bulk ^3He . By contrast, it will be shown here that the theory of the transverse relaxation for $x > 1.0$ depends crucially on the possibility of second-layer \vec{k} -state transitions upon exchange of first- and second-layer particles.

The first layer is taken to be a solid of N_1 particles in a triangular lattice with motion via quantum tunneling (at the rate corresponding to $x = 1$). The second layer is assumed to contain N_2 mo-

bile particles with second-layer coverage $x_2 \equiv x - 1 \cong N_2/N \ll 1$, with $N = N_1 + N_2$. A second-layer particle is presumed to tunnel rapidly from site to site, the sites being above the centers of the triangles established by the first-layer solid. There are thus $M = 2N_1$ sites, \vec{R}_a , on the second layer. A tight-binding wave function for a single particle in wave-vector and spin states (\vec{k}, μ) on the second layer is

$$\psi_{\vec{k}, \mu}(\vec{r}, \omega) = \frac{1}{\sqrt{M}} \sum_{\vec{R}_a} \exp(i\vec{k} \cdot \vec{R}_a) \varphi(\vec{r} - \vec{R}_a) \xi_{\mu}(\omega) \quad (2)$$

in which φ is a localized wave function and ξ_{μ} is a spin function.

The relaxation time T_2 is given in the Gaussian approximation for the correlation function, in terms of the second and fourth moments of the NMR line¹⁰ as

$$T_2^{-1} = (\pi/2)^{1/2} M_2 (M_2/M_4)^{1/2}. \quad (3)$$

The process we wish to consider here, namely interlayer exchange, alters M_4 so that it becomes $M_4^{(11)} + x_2 M_4^{(12)}$, where $M_4^{(11)}$ involves exchange within the first layer; and $M_4^{(12)}$, exchange between the layers. Calculations of M_2 and $M_4^{(11)}$ for the solid first layer have been reported previously.^{2,11} There are additional modifications on T_2 of the form $T_2^{-1} = (T_2^{-1})_{11} + x_2 (T_2^{-1})_{12} + x_2^2 (T_2^{-1})_{22}$, where $(T_2^{-1})_{nm}$ involves the dipolar interaction between particles in the n th and m th layers. The terms in x_2 and x_2^2 cause T_2 to decrease with increasing x_2 and so cannot provide an explanation of the data; however, their effect will be considered in a more comprehensive theoretical treatment which I do not describe here.

My starting point is the expression for the exchange-modulated fourth moment¹⁰

$$M_4 = -\frac{1}{2} \text{Tr} \{ \exp(-\beta H_L) [H_{\text{exch}}, [G_0, I_x]]^2 \} / \hbar^2 \text{Tr} [\exp(-\beta H_L) I_x^2], \quad (4)$$

in which $\exp(-\beta H_L)$ is the lattice density matrix (the high-temperature approximation has been made for the spin density matrix), H_{exch} is the exchange operator in Eq. (1), G_0 is the adiabatic portion of the dipolar Hamiltonian,¹⁰ and I_x is the total x component of spin. In the usual treatment¹⁰ of (4), the trace is just taken over the spin states, H_{exch} is an operator only in spin space, and the lattice is otherwise considered rigid. However, here one must consider the possibility of translational motion by the particles in the second layer, each having a wave function given by Eq. (2), with H_{exch} possibly having off-diagonal elements. Considering only pair exchanges, one has

$$\langle \vec{k}', i | H_{\text{exch}}(1, 2) | i, \vec{k} \rangle = -J_i(\vec{k}, \vec{k}') P_{12}^{(\sigma)}, \quad (5)$$

in which $J_i(\vec{k}, \vec{k}')$ is an exchange integral which is not diagonal in \vec{k} space. See Fig. 2(a). (Because the first layer and substrate can absorb or provide momentum, the second-layer \vec{k} state may change.¹²) $P_{12}^{(\sigma)}$ is the spin pair-permutation operator for particles 1 and 2 [$P_{12}^{(\sigma)} = \frac{1}{2}(1 + \vec{\sigma}_1 \cdot \vec{\sigma}_2)$], with 1 initially in the first layer, and 2 in the second. The density matrix is diagonal in the \vec{k} representation. For simplicity, the \vec{k} matrix elements of H_L are taken to be of the form $\epsilon_k = \hbar^2 k^2 / 2m^*$ all the way to the zone

boundary, where m^* is the effective mass for a particle in the band of second-layer states. Below I take $m^* = m$, the bare ^3He mass. Taking the spin trace results in

$$M_4^{(12)} = \frac{9}{32} \sum_{\vec{k}, \vec{k}'} \frac{\exp(-\beta\epsilon_{\vec{k}})}{Z} \sum_{i,j} (F_0^{ij})^2 [2J_i(\vec{k}, \vec{k}')J_i(\vec{k}', \vec{k}) + J_i(\vec{k}, \vec{k}')J_j(\vec{k}', \vec{k})], \quad (6)$$

in which i and j represent lattice sites on the first layer on which sit particles interacting by the spatial factor F_0^{ij} in the adiabatic portion G_0 of the dipolar Hamiltonian; Z is the density-matrix normalization factor.

One can transform Eq. (6) to a lattice representation by calculating $J_i(\vec{k}, \vec{k}')$ via the states of Eq. (2). One can show

$$J_i(\vec{k}, \vec{k}') = \frac{1}{M} \sum_{a,b} \exp(-i\vec{k} \cdot \vec{R}_a) \exp(i\vec{k}' \cdot \vec{R}_b) J_i(a, b), \quad (7)$$

in which $J_i(a, b)$ is an exchange integral representing a particle on second-layer site a hopping to first-layer site i , while the first-layer particle hops to second-layer site b [much as indicated in Fig. 2(a)]. Thus the localized form of exchange is also off-diagonal.¹³ The result is

$$M_4^{(12)} = \frac{9}{32} \sum_{\vec{k}, \vec{k}'} (F_0^{ij})^2 \sum_{a,b,c} \exp\left(-\frac{1}{2} \frac{m^*}{\beta\hbar^2} R_{ac}^2\right) \{2J_i(a, b)J_i(b, c) + J_i(a, b)J_j(b, c)\}. \quad (8)$$

Under the usual experimental conditions ($T = 1$ K), the density matrix in the localized representation [the exponential in Eq. (8)] is essentially unity for all relevant values of \vec{R}_{ac} in the lattice sum. Equation (8) can be considered in terms of diagrams showing the path of motion of the particles during the exchange process. Examples are shown in Figs. 2(b) and 2(c).

Equation (8) shows that, instead of a single exchange parameter, this theory has *several*, the values of which depend on the relative orientation, on the second-layer lattice, of \vec{R}_a and \vec{R}_b . I have not yet computed any of these parameters from first principles. Such a calculation requires correlated first-layer, second-layer pair wave functions. Preliminary work has shown that calculation of these wave functions presents unexpected difficulties.¹⁴ For the sake of a comparison with experiment I assume that all first-layer-second-

layer exchange integrals are identical and equal to the single parameter, J_{12} . The lattice sums may then be carried out and it is found, for the external field in the plane of the layers, that

$$M_4^{(12)} = 394.0 J_{12}^2 \hbar^2 \gamma^4 / d^6, \quad (9)$$

with d the nearest-neighbor distance in the first-layer solid and γ the gyromagnetic ratio. With $M_4^{(11)}$ taken from Ref. 2 or 11, I find

$$T_2 = (M_4^{(11)} + x_2 M_4^{(12)})^{1/2} / (\pi/2)^{1/2} M_2^{3/2} \\ \cong T_2(x_2 = 0) [1 + x_2 1.8 J_{12}^2 / J_{11}^2], \quad (10)$$

where J_{11} represents the exchange integral within the first-layer solid at monolayer completion. The experimental data² require $1.8 J_{12}^2 / J_{11}^2 \cong 17$, or $J_{12} / J_{11} \cong 3.0$. That this ratio is a reasonable one is shown crudely as follows: The separation between layers¹⁵ is ~ 3.4 Å and the nearest-neighbor distance at $x = 1$ is ~ 3.2 Å; then from Ref. 2, one obtains $J_{11}(d = 3.4) / J_{11}(d = 3.2) \cong 3.5$.

The results of Eq. (8) can be derived directly by taking the trace in localized states, however; then the density matrix $\exp(-\beta H_L)$ is also non-diagonal because H_L includes particle hopping in the second layer. Its matrix elements [as in Eq. (8)] can be found via the \vec{k} states which diagonalize it.

The important feature of the above explanation is the need for off-diagonal elements of H_{exch} in \vec{k} space. If one tries to make use of a Heisenberg-type Hamiltonian [e.g., Eq. (5)] with no off-diagonal elements, one finds an $M_4^{(12)}$ of order $1/N$, which is obviously meaningless.

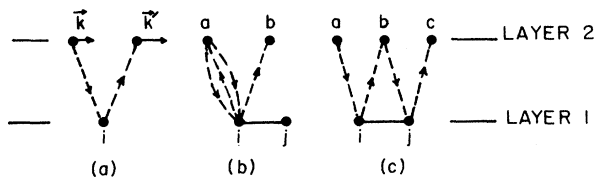


FIG. 2. (a) A representation of the exchange-operator matrix element $J_i(\vec{k}, \vec{k}')$ showing two particles interchanging layers and the change of \vec{k} states. The particle in state \vec{k} drops in the first layer while the particle on site i jumps to the second layer and into state \vec{k}' . (b), (c) Typical diagrams occurring in the lattice sums of Eq. (8). The line connecting i and j in the first layer represents the dipolar interaction.

Some interesting possibilities concerning future experiments present themselves. If x_2 is large enough and the temperature low enough, the Fermi fluid of the second layer may approach degeneracy which will limit the scattering from \vec{k} to \vec{k}' and may give a characteristic temperature dependence to the process. However, for large x_2 , effects of order higher in x_2 than the linear ones considered here may also become important. T_2 may also show the effects of second-layer solidification as the third layer begins to form. These effects will be studied theoretically in future work.

This research was supported in part by the National Science Foundation under Grant No. DMR-7614447.

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¹⁰What is needed here is the second time derivative of the spin-torque correlation function. I have been able to show that the derivative is given by Eq. (4) and is equal to the fourth moment of the resonance line. Equation (4) is an extension of the results given by R. C. Richardson, A. Landesman, E. Hunt, and H. Meyer [*Phys. Rev.* **146**, 244 (1966)], and by A. Landesman [*Ann. Phys. (Paris)* **8**, 53 (1973)].

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¹²Cf. Ref. 7, Appendix A.

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¹⁵Novaco, Ref. 14.

Superfluid Transition of ^4He Films Adsorbed on Porous Vycor Glass

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(Received 18 April 1977)

The temperature variation of the superfluid density has been measured for ^4He films adsorbed on porous Vycor glass. Films with transition temperatures ranging from 0.1 to 1.5 K were studied. Apart from a small rounding on the order of $5 \times 10^{-3} T_c$, the superfluid density of these films follows a power law near the transition similar to bulk helium.

Experimental studies of the superfluid transition in thin films of adsorbed ^4He atoms have promised to provide an excellent opportunity to observe the influence of size and dimensionality on a phase transition. The ease with which the film thickness can be altered and the wide range of substrates of varying topology and surface condition should make the helium film an extremely versatile system. Unfortunately, until recently there have been few experiments with the necessary precision required for critical-point studies.

In fact, the nature of the superfluid phase transition in thin ^4He films has remained an open and controversial question.¹⁻³

The principal difficulty has been the lack of experimental probes of sufficient sensitivity to examine the superfluid properties in the immediate vicinity of the superfluid onset or transition temperature. Prior to the present work, the only method which provided a measure of the superfluid mass through the region of the transition was the quartz microbalance technique developed