Dupree[1] obtained a simple expression for the resonance width which is in agreement with the result here.[12]

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[12] However, the method here of introducing the resonance widths into the weak-turbulence equations (i.e., by smoothing over both waves and particles) is not in agreement with that of Dupree. This can be traced to Dupree's neglect of the effect of his operator on $f_0(v)$ which probably should be retained.

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# Four-Particle Exchange in Solid $^3$He

A. K. McMahan* and J. W. Wilkins†
*Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14853*

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We report calculations which suggest that there is a physically important four-atom exchange process in bcc $^3$He and thus an important four-spin term in the exchange Hamiltonian. A simple, mean-field analysis of this Hamiltonian appears to account for a number of the perplexing properties of bcc $^3$He. An understanding of other properties may require treatment of the exact four-spin term. It is our hope to stimulate such effort by this Letter.

We report the results of calculations which suggest there is a physically important four-atom exchange process in bcc solid $^3$He. The process gives rise to a *four-spin* term in the effective spin or exchange Hamiltonian with an exchange energy comparable to the nearest-neighbor two-spin term. A simple-minded mean-field treatment suggests that this four-spin term could lead to a temperature-dependent exchange frequency which offers partial insight to the several perplexing properties of bcc solid $^3$He.[1]

To facilitate discussion we define the exchange Hamiltonian including pair, triple, and the important cyclic quadruple exchange[2]:

$$H_{\text{ex}} = -2[J_1 - 6J_{112} + 3J_{1111,23}] \sum_{i<j}^{(1)} \vec{I}_i \cdot \vec{I}_j - 2[J_2 - 4J_{112} + J_{1111,23}] \sum_{i<j}^{(1)} \vec{I}_i \cdot \vec{I}_j - 4J_{1111,23} \sum_{i<j<k<l} [(\vec{I}_i \cdot \vec{I}_j)(\vec{I}_k \cdot \vec{I}_l) + (\vec{I}_j \cdot \vec{I}_k)(\vec{I}_l \cdot \vec{I}_i) - (\vec{I}_i \cdot \vec{I}_k)(\vec{I}_j \cdot \vec{I}_l)]. \tag{1}$$

The first two-spin term involves nearest-neighbor spins [the (1) over the sum], while the second involves next-nearest-neighbor spins [the (2)]. Finally the one four-spin term involves four atoms located at the corners of the rhombus, lying in the (110) plane, whose sides $\overline{ij}$, $\overline{jk}$, $\overline{kl}$, and $\overline{li}$ are first-neighbor distances (the subscript 1111 in $J_{1111,23}$) and whose diagonals $\overline{ik}$ and $\overline{jl}$ are second- and third-nearest neighbors (the 23).

Later in this Letter we will present a summary of the calculation for the various exchange constants. First we want to briefly discuss the anomalies in $^3$He solid within the context of a mean-field approximation for (1) in which, for simplicity, we ignore the next-nearest-neighbor exchange terms except to indicate those situations where they may play a noticeable role. Then (1) reduces to

$$H = -2\tilde{J}_1(T) \sum_{i<j}^{(1)} \vec{I}_i \cdot \vec{I}_j, \qquad (2)$$

where

$$\tilde{J}_1(T) = J_1 - 6J_{112} + 3J_{1111,23}(1 + 4\langle \vec{I}\cdot\vec{I}\rangle_1). \qquad (3)$$

In (3) the term $\langle \vec{I}\cdot\vec{I}\rangle_1$ corresponds to the thermal expectation value of the scalar product of nearest-neighbor spins. Since we expect $^3$He solid to become antiferromagnetic at low temperatures, then $\langle \vec{I}\cdot\vec{I}\rangle_1$ will change from zero at high temperature to something like $-\frac{3}{4}$ in the ordered phase. Taking a simple analytic form for this switching behavior, and using numbers we have calculated, we might approximate (3) above the transition by

$$\tilde{J}_1(T) \approx -0.65 + 0.4 \tanh(2/T), \qquad (4)$$

where $\tilde{J}_1$ and $T$ are measured in millikelvins. [The factor 2 in the $\tanh(2/T)$ is an estimate of the transition temperature in the absence of $J_{1111,23}$.]

There are several puzzling anomalies in the $^3$He solid data which we discuss in terms of (2) and (4). We restrict ourselves to a single density, corresponding to a molar volume of 24.0 cm$^3$/mole.

*High-temperature results* ($T > 20$ *mK*).—In this regime, $\tilde{J}_1$ is a temperature-independent constant. High-temperature susceptibility measurements[3] indicate it is negative (antiferromagnetic), with its best value of $-0.65$ mK coming from the pressure measurements.[4] When the pressure measurements are extended to finite magnetic fields,[5] it appears necessary to assume an additional ferromagnetic next-nearest-neighbor interaction of strength $\tilde{J}_2/\tilde{J}_1 \sim -0.2$.[6,7] All of the *high*-temperature data can be reconciled within such a two-parameter model, in which $\tilde{J}_2$ is ferromagnetic.[6,7] Ratios in the vicinity of $-0.2$ have been theoretically calculated.[6,8,2] In the present work we can only specify $+0.03 > \tilde{J}_2/\tilde{J}_1 > -0.2$ because of 10% or so uncertainties in the various exchange frequencies, and large cancelations in $J_2 - 4J_{112} + J_{1111,23}$ [the high-temperature value of $\tilde{J}_2(T)$].

*Low-temperature results* ($T < 20$ *mK*).—(i) transition temperature. A constant first-neighbor exchange frequency of $-0.65$ mK corresponds (via $T_\mathrm{N} = 2.75|J|$) to an antiferromagnetic transition temperature of 1.8 mK (a bit higher if $\tilde{J}_2 > 0$), whereas the first observed transition in the solid is 1.1 mK.[9] Clearly (4) would produce a lower transition temperature. Based on its low-temperature limit, we get 0.7 mK. But we would stress that the results here and below, being sensitive to the guessed interpolation for $\tilde{J}_1(T)$, are at best semiquantitative.

(ii) Specific heat. A high-temperature expansion for the specific heat in powers of $T^{-1}$ with use of an antiferromagnetic Heisenberg Hamiltonian results in a $T^{-3}$ term with a positive coefficient (even larger if $\tilde{J}_2 > 0$); whereas the experiments[9,10] indicate a *negative* one. A temperature-dependent exchange such as (4) can result in a negative coefficient, although our crude argument gives a coefficient a factor of 2 too small. Near the transition temperature of the solid the specific heat has a pronounced bump[9,10] near 2 mK prior to the entropy anomaly at 1.1 mK. Our mean-field model *cannot* explain this structure. On the other hand, the original Hamiltonian (1) we propose may be able to explain it. Specifically we propose to the phase-transition-theory community the following question: Does $H_\mathrm{ex}$ (1) with its four-spin term have the same behavior as a Heisenberg Hamiltonian in the vicinity of the (depressed) transition temperature? Or can it exhibit structure consistent with that observed in bcc $^3$He?

(iii) Spin diffusion. As additional evidence that something extraordinary is occurring at about 2 mK in the solid, there is the fact that the spin diffusion coefficient $D$ drops sharply by over 30% for decreasing temperature about 2 mK,[11] which leads us once again to the idea that the effective $J$ ($\propto D$) is strongly temperature dependent.

(iv) $H$-$T$ phase boundary. In terms of a mean-field model, an external magnetic field would favor a positive $\langle \vec{I}\cdot\vec{I}\rangle_1$ in (3) and hence increase $|\tilde{J}_1|$. Presumably this effect would increase the magnetic field at which the spin-flop to paramagnetic-solid transition occurs over that predicted by a near-neighbor Heisenberg Hamiltonian consistent with the observed spin-ordering temperature. Only limited experimental data[12] exist for the $H$-$T$ phase boundary.

Finally we turn to the actual calculation of $J_{1111,23}$ and the other exchange frequencies. There is an extensive literature[13] which underpins our calculation. The exchange frequencies can be written in terms of integrals involving the Jastrow-Gaussian *Ansatz* for the many-body wave function in solid $^3$He:

$$\psi = \prod_i \exp[-\tfrac{1}{2}A(\vec{r}_i - \vec{R}_i)^2] \prod_{i<j} f(r_{ij}), \tag{5}$$

where $\vec{r}_i$ and $\vec{R}_i$ specify the atom and lattice site positions, respectively.

The scalar coefficient $A$ is proportional to the (Einstein) frequency describing the harmonic motion of an atom about its lattice site. In our calculation we generalize $A$ to a matrix[14] in order to take into account a more realistic description of the atomic motion based on a self-consistent calculation[15] of the phonon frequencies. Accordingly the $A_{\text{eff}}$ listed in Table I for the four largest exchange frequencies reflects phonon frequencies which predominate in that exchange process.

The Jastrow function $f(r_{ij})$ is roughly a step function used to describe the short-range correlations, by preventing two atoms from approaching closer than their hard-core diameter. The energy of solid He can be written in terms of integrals involving $\psi$. Various calculations[16] which in effect minimize the energy with respect to functional form and parameters of (5), yield very similar Jastrow functions. On the other hand the energy is not very sensitive to $A$. Nevertheless, it now appears on the basis of recent Monte Carlo calculations,[8] that $A$ may be 30% smaller than the range of values previously accepted. We exploit this possibility in our work.

In terms of the wave function (5) the exchange frequency for a given exchange $p$ of atoms is

$$J_p = -\Omega_p \exp(-A d_p^{\,2}/4) I_p(A, \{f\}), \tag{6}$$

where $d_p$ is a distance in configuration space between the original and exchanged atoms. Values of $d_p$ in units of the nearest-neighbor distance $R$ are given for various exchanges in Table I. The attempt frequency for the exchange process, within the Einstein approximation for the atomic motion, is given by $\Omega_p = (\hbar/2M)d_p(A^3/\pi)^{1/2}$.

The exchange frequencies are dominated by the other two factors in (6). The first factor is called the Gaussian overlap. The exponential factor is a measure of the overlap of the wave functions of the original and exchanged atoms. Note that it is a very sensitive function of the phonon frequencies appropriate to a given exchange, as is indicated by the values of $A_{\text{eff}}$ in Table I. The seemingly small difference between $A_{\text{eff}}$ for the quadrupole and near-neighbor-pair cases results in nearly an order of magnitude increase in the ratio $J_{1111,23}/J_1$ over what its value would be in the Einstein approximation, where all the $A_{\text{eff}}$ would be a single value.

The second factor is related to hard-core effects. The final factor $I_p$ corrects for the fact that the Gaussian overlap overestimates the probability for atoms to exchange by not excluding routes of exchange forbidden by the hard-core repulsion of the atoms. This effect is most important for pair exchange where straight-line exchange would have the atoms sitting on top of each other. In this case $I_1$ includes the dominant factor $\exp(-\frac{1}{2}A\sigma^2) \sim \frac{1}{25}$, where $\sigma$ is the hard-core radius. This strong dependence on $A$ does *not* occur in $I_p$ for three- and four-atom exchange, where the short-range correlation effects are largely geometric in nature.[2] The values for $I_p$ in Table I are extrapolated from the results of Ref. 2 based on Monte Carlo integrations (for the Einstein case), with the exception of the last (and crucial) entry which has been estimated on the basis of the experience gained from three-atom exchange.

In the next to the last column of Table I are the results for $J_p$. It is clear that $J_{1111,23}$ is too small *but* so is $J_1$ by at least a factor of 20, if it is to agree with the high-temperature measure-

TABLE I. Exchange frequencies and related intermediate numbers for bcc $^3$He (24.0 cm$^3$/mole).

| | $(d_p/2R)^2$ | $A_{\text{eff}}$ (Å$^{-2}$) | $\Omega_p \exp(-A_{\text{eff}} d_p^{\,2}/4)$ | $I_p$ | $J_p$ (mK) | Scaled $J_p$ (mK) |
|---|---|---|---|---|---|---|
| Pair (1) | 1/2 | 1.350 | 2.00 | 0.014 | −0.028 | −1.02 |
| (2) | 2/3 | 1.129 | 0.602 | 0.0067 | −0.0040 | −0.25 |
| Triple (112) | 5/6 | 1.295 | 0.0104 | 0.086 | −0.00089 | −0.085 |
| Quadruple (1111, 23) | 1 | 1.136 | 0.0042 | 0.09 | −0.00038 | −0.045 |

ments. At this stage we take advantage of the fact that the more accurate Monte Carlo work[8] suggests that the true $A$ values may be 30% or so smaller than those consistent with Ref. 15 on which our work is based. Accordingly we have decreased $A$, or more accurately, rescaled downward the phonon frequencies by 32% so that $\tilde{J}_1(T=\infty)$ [see (3)] agrees with the value $-0.65$ mK deduced from the high-temperature, zero-field, pressure measurements.[4] The resulting numbers are shown in the last column, and have been used in arriving at the various numerical values in our paper [e.g., $9|J_{1111,23}|$ is 0.4 mK, the factor used in (4)]. We emphasize that $\tilde{J}_1(T=\infty)$, $J_{1111,23}$, and $\tilde{J}_2(T=\infty)$ are only weakly related, and so our conclusions about the last two quantities are not compromised by our procedure of fitting the first. We also emphasize that the $A$ value consistent with our rescaled phonon frequencies (those used in calculating the last column of Table I) is in fact in good agreement with that of Ref. 8. As a result of these calculations we argue that this four-atom exchange (the next largest one is an order of magnitude smaller) must be taken into account in any treatment of the phase transition of bcc $^3$He.

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