

are approximated by $\delta v_{\perp} \approx \alpha_{\perp}/(4j)^{1/2}$. They are obtained by successive differentiations of $F_0^{(0)} = (1/\pi\alpha_{\perp}^2) \exp(-v_{\perp}^2/\alpha_{\perp}^2)$ with respect to α_{\perp}^2 . Thus, the integration of Eq. (2) is readily performed. We find the following: (1) For $j=0, 1, 2$, the distributions are stable with respect to $k_{\parallel}=0$ waves. (2) For distributions with $j=3, 4, 5, \dots$, the zero-frequency mode is unstable. The dependence of threshold density on relative half-width is shown in Fig. 1. Those waves are unstable whose propagation vectors fall in the band $2.5 \lesssim k\rho_g \lesssim 3.8$, where ρ_g is the gyroradius corresponding to the peak value of v_{\perp} . (3) The $j=6, 7, \dots$ distributions sustain a growing wave whose real component of frequency is $\approx 1.2\omega_c$, with density threshold given by $\omega_p \approx 10\omega_c$ and $3.8 \lesssim k\rho_g \lesssim 5.0$. (4) As j increases, the results go over smoothly to those obtained with the distribution of Eq. (3). That is, higher frequency modes appear for threshold densities and bands of k which tend smoothly to those given by the distribution function of Eq. (3). We have observed unstable growth rates, dependent upon the density excess above the threshold value, which are typically some tenths of the gyrofrequency for excesses of the order of 10% of the threshold value.

The point to be emphasized here is that $k_{\parallel}=0$ modes can be stabilized by a moderate amount

of broadening and smoothing of an initially sharply peaked distribution. The absence of particles with small v_{\perp}^2 does not have to be eliminated completely.

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STUDY OF THE EXCHANGE INTEGRAL OF CRYSTALLINE ³He AT 0°C

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The nuclear magnetic properties of crystalline ³He have been studied extensively in recent years.¹ In particular, there have been many experimental²⁻¹⁰ and theoretical¹¹⁻¹⁴ investigations aimed at obtaining an accurate value of the exchange integral J . All calculations of J have taken the effects of pair correlations into account. Bernardes and Primakoff¹¹ included them in a phenomenological way, whereas Saunders¹² derived an approximate differential equation for the correlation function. Recently Garwin and Landesman¹⁴ have calculated J by means of an extension of Saunders's work.

The purpose of this note is to extend recent

calculations of the ground-state energy E_0 ^{15,16} to include the effects of exchange. The cluster expansion of E_0 used previously is generalized so that properly symmetrized wave functions may be treated. With an antisymmetrized version of the Jastrow-type wave function used previously, an expression is obtained for J which takes the effect of pair correlations into account in a systematic way. Calculations of J as a function of the nearest-neighbor distance are presented for both the bcc and hcp structures of crystalline ³He. The effects of the pair correlations on J are analyzed.

The cluster development of the energy can

be made readily using van Kampen's approach.¹⁷ One finds

$$E_0 = \sum_{i=1}^N \mathcal{E}_1(i) + \sum_{1 \leq i < j \leq N} [\mathcal{E}_2(i, j) - \mathcal{E}_1(i) - \mathcal{E}_1(j)] + \dots, \quad (1)$$

where \mathcal{E}_1 and \mathcal{E}_2 (one- and two-particle energies) are given by

$$\mathcal{E}_1(i) = \langle \psi_1(i) | H_1(i) | \psi_1(i) \rangle / \langle \psi_1(i) | \psi_1(i) \rangle; \quad (2)$$

$$\mathcal{E}_2(i, j) = \langle \psi_2(i, j) | H_2(i, j) | \psi_2(i, j) \rangle / \langle \psi_2(i, j) | \psi_2(i, j) \rangle. \quad (3)$$

Here $H_1(i)$ and $H_2(i, j)$ are appropriate one- and two-particle Hamiltonians which we take to be

$$\begin{aligned} H_1(i) &= -(\hbar^2/2m)\nabla_i^2, \\ H_2(i, j) &= H_1(i) + H_1(j) + v_{ij}, \end{aligned} \quad (4)$$

where v_{ij} is the pair interaction. Further, $\psi_1(i)$ and $\psi_2(i, j)$ are appropriate one- and two-body wave functions which, in general, would be obtained in a self-consistent way and would, therefore, not be eigenfunctions of $H_1(i)$ and $H_2(i, j)$. The higher terms of (1) can be constructed straightforwardly, but will not be considered explicitly here.

Following previous work,^{15,16} we now introduce the single-particle function $\varphi_j(i) = \varphi(|\vec{r}_i - \vec{R}_j|)$ and the correlation function $f_{ij} = f(|\vec{r}_i - \vec{r}_j|)$; \vec{r}_i and \vec{R}_j are, respectively, the coordinates of the i th particle and j th lattice site.

Further, we choose

$$\psi_1(i) = \varphi_i(i) s_i, \quad (5)$$

$$\psi_2(i, j) = [\varphi_i(i)\varphi_j(j) \pm \varphi_j(i)\varphi_i(j)] f_{ij} s_{ij}, \quad (6)$$

where s_i and s_{ij} are one- and two-particle spin functions. We make this choice of wave functions to obtain a small contribution from the higher terms of (1).¹⁵ Since ³He has a nuclear spin of $\frac{1}{2}$, the plus and minus signs in (6) go with singlet and triplet spin states, respectively.

We may now write \mathcal{E}_1 and \mathcal{E}_2 more explicitly by making appropriate partial integrations^{18,19} (the spin sums cancel). We find

$$\mathcal{E}_1(i) = \langle P_{ii} \rangle, \quad (7)$$

$$\mathcal{E}_2(i, j) = K_{ij} \pm \frac{1}{4} J_{ij}, \quad (8)$$

where

$$K_{ij} = [\langle f_{ij}^2 \rangle - \langle f_{ij}^2 \rangle_x]^{-1} [\langle f_{ij}^2 (P_{ii} + P_{jj} + V_{ij}) \rangle \langle f_{ij}^2 \rangle + \langle f_{ij}^2 (P_{ij} + P_{ji} + V_{ij}) \rangle_x \langle f_{ij}^2 \rangle_x] - \langle P_{ii} \rangle - \langle P_{jj} \rangle, \quad (9)$$

$$J_{ij} = 4[\langle f_{ij}^2 \rangle - \langle f_{ij}^2 \rangle_x]^{-1} [\langle f_{ij}^2 (P_{ij} + P_{ji} + V_{ij}) \rangle_x \langle f_{ij}^2 \rangle - \langle f_{ij}^2 (P_{ii} + P_{jj} + V_{ij}) \rangle \langle f_{ij}^2 \rangle_x]. \quad (10)$$

In (7), (9), and (10),

$$P_{ij} \equiv [-\hbar^2/8m \varphi_j(i) \varphi_i(i)] [\varphi_j(i) \nabla_i^2 \varphi_i(i) + \varphi_i(i) \nabla_i^2 \varphi_j(i) - 2\vec{\nabla}_i \varphi_j(i) \cdot \vec{\nabla}_i \varphi_i(i)], \quad (11)$$

$$V_{ij} \equiv v_{ij} + (-\hbar^2/2m f_{ij}^2) (f_{ij} - \nabla_i^2 f_{ij} - \vec{\nabla}_i f_{ij} \cdot \vec{\nabla}_i f_{ij}), \quad (12)$$

and the averages $\langle \rangle$ and $\langle \rangle_x$ are taken with respect to the weight functions $w^{-2} \varphi_i^2(i) \varphi_j^2(j)$ and $w^{-2} \varphi_i(i) \varphi_j(j) \varphi_i(j) \varphi_j(i)$, respectively; $w = \int d\vec{r} \varphi^2(r)$. When the effects of exchange vanish, (9) reduces to the result obtained previously.¹⁵ When $f_{ij}^2 \rightarrow 1$, (10) reduces to the results of Carr²⁰ and Mullin.²¹

The factor of 4 has been introduced in (10) so that the difference in energy between a parallel and antiparallel alignment of spins is $\frac{1}{2} J_{ij}$; this definition agrees with that of GL. The definition is such that $J_{ij} > 0$ implies a ferromagnetic and $J_{ij} < 0$ implies an antiferromag-

netic ground state. The spin-alignment temperature T_c is then

$$T_c \approx \frac{1}{4} Z |J|, \quad (13)$$

where Z is the number of nearest neighbors and J is the exchange integral for nearest neighbors. It should be pointed out that there are contributions to J from the higher terms of (1); however, on physical grounds, they are quite small.

To calculate J we used the analytic forms $\varphi(r) = \exp(-\frac{1}{2}Ar^2)$ and $f(r) = \exp\{-K[(\sigma/r)^\nu - (\sigma/r)^6]\}$. Here σ is the collision diameter (2.556 Å for He), and A , K , and ν are variational parameters determined to minimize E_0 .¹⁶ With these choices, we have

$$J = -\left[\frac{\hbar^2 A^2 R^2}{m} + 4 \frac{\langle f^2 V \rangle}{\langle f^2 \rangle} \right] \frac{\langle f^2 \rangle_x}{\langle f^2 \rangle} + 4 \frac{\langle f^2 V \rangle_x}{\langle f^2 \rangle}, \quad (14)$$

where we have dropped subscripts since everything refers to nearest neighbors (R is the nearest-neighbor distance). It turns out that the first term in brackets in (14) is about three orders of magnitude larger than the second and relatively insensitive to variations in R . Thus J is most sensitive to $\langle f^2 \rangle_x$ and $\langle f^2 V \rangle_x$.

We have calculated T_c as a function of R using $\nu = 12$. The results are plotted in Fig. 1. The tolerances used in determining A and K were such that J is numerically accurate to 10%. Our values of T_c are in reasonably good agreement with experiment, although the dependence on R is quite different. It is our opinion that this behavior (i.e., values of J which are too low for large R and which decrease too slowly as R decreases) is a consequence of the choice of a Gaussian $\varphi(r)$. In the first place, even when R is large and the "correct" $\varphi(r)$ is well approximated by a Gaussian for $r \lesssim \frac{1}{2}(R-\sigma)$, the correct $\varphi(r)$ will certainly be larger than the Gaussian for $r \gtrsim \frac{1}{2}(R-\sigma)$ because the "effective" potential well¹⁶ levels off at about this distance. Hence, we obtain a too-small J for large R . In the second place, the "correct" $\varphi(r)$ should be more and more like a square-well wave function as R decreases. The Gaussian $\varphi(r)$ cannot give this behavior, and will therefore yield a $J(R)$ which does not decrease fast enough as R increases.

We have also investigated the sensitivity of J to $f(r)$. We varied K from 0.14 to 0.18 and found that J varied by a factor of 3 while E_0

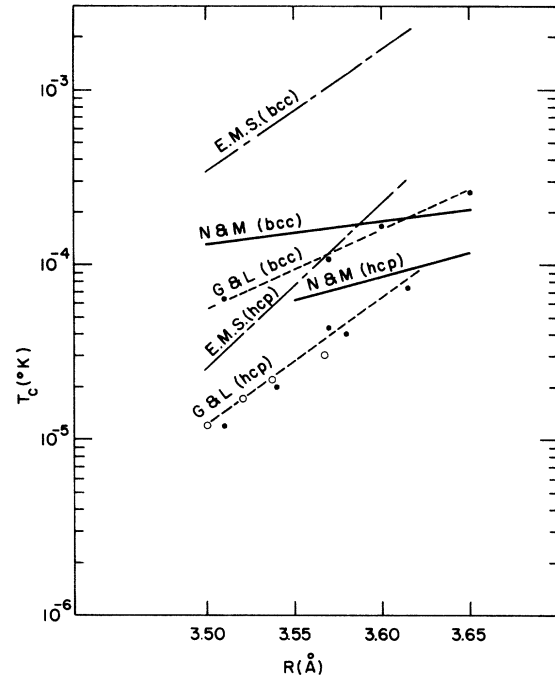


FIG. 1. The spin-alignment temperature T_c as a function of R , the nearest-neighbor distance. The solid lines are the results of our calculations. The rest of the graph is taken from Fig. 6 of GL. The center lines are Saunders's results as corrected by GL. The open and solid circles are values of T_c calculated by GL from experimental measurements of GL and Reich.⁷

only changed by 1%. We also varied ν from 12 to 8 and found that J changed by a factor of 4 while E_0 changed by 10%. Furthermore, J (i.e., the integrals $\langle f^2 \rangle_x$ and $\langle f^2 V \rangle_x$) is most sensitive to $\varphi(r)$ for $\frac{1}{2}(R-\sigma) \lesssim r \lesssim \frac{1}{2}(R+\sigma)$ and to $f(r)$ for $0 \lesssim r \lesssim \frac{1}{2}\sigma$, whereas E_0 is most sensitive to $\varphi(r)$ for $0 \lesssim r \lesssim \frac{1}{2}(R-\sigma)$ and to $f(r)$ for $\frac{1}{2}\sigma \lesssim r \lesssim \frac{3}{2}\sigma$. Since J and E_0 are most sensitive to parts of $\varphi(r)$ and $f(r)$ which overlap only slightly, this "overlap" region would have to be important in the variational calculation of E_0 in order to ensure the correctness of J . Due to the difficulty of assessing the importance of this "overlap region" in our calculations, we must point out that the reliability of our values of J is uncertain.

In our calculations we have always found that J is negative. However, we also want to point out that the first and third terms of (14), which contribute most to J , are nearly equal. E.g., at $R = 3.55$ Å in the bcc phase, these terms are -5.24×10^{-4} and $+4.92 \times 10^{-4}$ cal/mole, respectively. Thus, our present calculations also

leave an uncertainty in the sign of J . In addition, there is the possibility that J could change sign as R decreases further, in line with the prediction of BP, although our calculations do not indicate this behavior.

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ENERGY TRANSFER FROM COPPER AND SILVER TO RARE EARTHS IN II-VI COMPOUNDS

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We have found that rare-earth fluorescence can be excited by energy transfer from copper and silver in ZnS, CdS, and ZnSe. No report of a similar sensitization has been made by previous investigators of the fluorescence of rare-earth-doped ZnS.¹⁻⁴ A specific example of this system is ZnS:Cu, Er, whose strongest group of lines near 18 100 cm⁻¹ corresponds to the transition ⁴S_{3/2} - ⁴I_{15/2} of the free Er ion. The excitation spectra of these lines, for samples of widely varying Cu content, are shown in Fig. 1. The enhancement produced by the addition of copper is clearly significant, and even the high-purity fluorescent-grade ZnS used in these experiments apparently contains enough Cu (about 0.3 ppm) to make its presence obvious.

Line emission has been observed in ZnS:Cu, Ln for Ln = Pr, Nd, Tb, Dy, Ho, Er, and Tm. Fluorescence generally occurs from several levels for each ion, but except for Tm the strongest emission occurs from levels between 18 000

and 21 000 cm⁻¹ above the ground state. No line emission has been similarly excited when Ln = Sm, Eu, and Yb. The excitation and emission spectra indicate that these three ions are reduced to the divalent state in our samples.

The tendency for all of the lanthanides in ZnS:Cu, Ln to emit from levels near 20 000 cm⁻¹ indicates that the sensitization may involve a resonance transfer from an energy level of a Cu defect at or slightly above 20 000 cm⁻¹. To determine if such a level is present we have studied the spectra of ZnS:Cu, Gd. No energy transfer can occur to Gd since its lowest excited states lie above 32 000 cm⁻¹ (4.0 eV), which is greater than the band gap of ZnS. The emission of ZnS:Cu, Gd is thus likely to be characteristic of the Cu sensitizing defect. This emission is a broad band which maximizes at 23 000 cm⁻¹ (2.86 eV) and decreases to 10% intensity at 20 800 and 24 900 cm⁻¹. The excitation spectrum of this band is identical to that of the line emission of ZnS: