# Calculation of exchange integrals in solid ³He

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The first two terms in the expansion of the logarithm of the exchange integral in powers of the lattice constant of a quantum ³He crystal are calculated in the semiclassical approximation. Numerical calculations are carried out for the exchange of two, three, and four neighboring atoms.

In this letter we shall carry out a microscopic calculation of certain exchange integrals in order to determine how many-particle exchange processes[1] affect the ³He Hamiltonian. The method of Ref. 2 is used to calculate the exchange integrals for three processes: the exchange of two nearest neighbors, $J_{nn}$; the exchange of three atoms, $J_t$; and the planar exchange of four atoms in the bcc phase of ³He, $K_p$;

For the calculations we use a semiclassical approximation of $\Psi(X)$, the wave function of the $N$-atom quantum crystal, where $X$ is a $3N$-dimensional vector specifying the coordinates of all the atoms.[2] The exchange process in a quantum crystal may be summarized as follows: A system which is initially near the point $X = X_1$, which is a minimum of the potential energy $U(X)$, undergoes a transition (tunnels) to the point $X_2$, which is another minimum of the potential energy. The semiclassical approximation requires calculation of the tunneling trajectory $X(t)$ from $X_1$ to $X_2$ from the principle of least action. The point $X_2$ differs from $X_1$ in that the coordinates of two, three, or four atoms have been interchanged, but in a calculation along the tunneling trajectory it is necessary to consider the displacements of other lattice

atoms also. In other words, the problem of calculating the tunneling trajectory is a many-body problem.

Another difficulty is that in $^3$He the potential energy $U(X)$ cannot be written as the simple sum of the binary interaction potential $v(r)$ over all pairs of atoms. The contribution of each pair to $U(X)$ must be averaged over the zero-point vibrations. For the potential energy we use the approximation

$$U(X) = \frac{M}{2} \sum_k \omega^2(k) \zeta^2(k) + U_a(X). \tag{1}$$

The first term is the lattice energy in the harmonic approximation; the sum is over the variable $k$, which includes the wave vector and the polarization of the phonon; and $\zeta(k)$ and $\omega(k)$ are the amplitude and frequency of the phonons. In calculating $\omega(k)$ we use the approximation of an elastic and isotropic medium (the Debye model). The velocity of sound is determined from experimental data for the Debye temperature and the elastic moduli.[3] The second term in (1) is an anharmonic increment $U_a(X)$. It is important to note that $U_a$ is determined solely by a sum over a small number of atoms—those for which the displacement cannot be assumed small. We use an interpolation formula in the calculation of $U_a$. In this procedure the contribution of a particular pair of atoms to $U_a$ is zero if the interatomic distance is approximately equal to the equilibrium value, while the contribution is equal to the repulsive part of the Lennard-Jones potential if the atoms move very close together.

In the calculation of the tunneling trajectory $X(t)$ the effective duration of the anharmonic effects, $\tau$, is small in comparison with the characteristic times for harmonic oscillations, $\omega_D \tau \ll 1$, because of the rapid increase in the interatomic potential as the atoms move close together. Consequently, a perturbation for the atomic displacement does not manage to propagate through the crystal until the tunneling trajectory intersects the point $t = 0$, which is chosen halfway along the trajectory. The tunneling trajectory is sought by a "firing" method. Two trajectories $X(t)$ and $X'(t)$ are chosen in such a manner that at $t = -\infty$ we have $X(t) = X_1$ and $X'(t) = X_2$. At the time $t = 0$ the two trajectories join smoothly:

$$X(0) = X'(0), \qquad \dot{X}(0) = -\dot{X}'(0). \tag{2}$$

The "firing" process can be arranged in such a manner that joining condition (2) is checked for only those atoms for which the anharmonic effects must be taken into account.

We can transform to dimensionless displacements in (1) by expressing distances in the units of the lattice constant $a$ (in the bcc crystal we label an edge of the cube as $a$). If the dimensionless displacement is fixed, then $U_a$ in (1), which is determined by the repulsive part of the Lennard-Jones potential, varies $\propto a^{-12}$. Experiments show that the characteristic frequencies in helium vary approximately $\propto a^{-7}$, so that the harmonic term in (1) also varies $\propto a^{-12}$. It is thus possible to calculate the tunneling trajectory and the action along this trajectory for only a single value of $a$ and then scale the results ot other densities. Using the results of Ref. 2, we can write the following expression for the exchange integral:

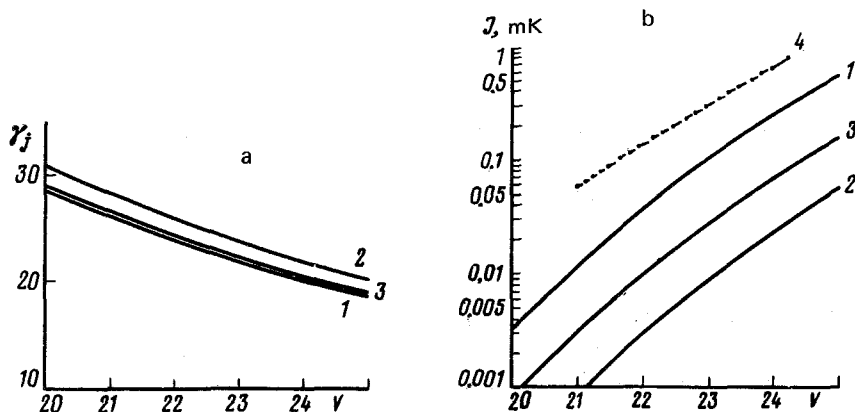$$\ln|J| = -S_0 \gamma_0^{1/2} (\sigma/a)^5 - 9,5 \ln a + \ln A_0. \tag{3}$$

FIG. 1. a—The Heisenberg constant $\gamma_j$; b—exchange integrals, as functions of the molar volume $V$ (in cubic centimeters per mole). Curves 1, 2, and 3 correspond to binary, ternary, and quaternary exchange, respectively; curve 4 represents the experimental data.[3]

The parameter $\gamma_0 = 4\epsilon M(\sigma l/\hbar)^2$, which arises in the transformation to dimensionless units ($h = a = M = 1$), is a measure of the quantum properties of the crystal; $M = 5 \times 10^{-24}$ g; and $\epsilon = 10.2$ K, $\sigma = 2.56$ Å, and $l = 12$ are the parameters of the repulsive part of the Lennard-Jones potential. Numerical calculations yield values of 3.81, 4.08, and 3.86 for $S_0$ for the exchange of two, three, and four atoms, respectively.

Figure 1a shows the magnetic Grüneisen constant $\gamma_j = \partial(\ln|J|)/\partial(\ln V)$ for the three exchange processes as a function of the molar volume $V$. The Grüneisen constant, determined from measurements of the helium pressure in a magnetic field,[3] varies from 17.5 to to 19.2 as the volume changes from 24 to 22 cm$^3$/mole. If we assume that the thermodynamic properties are determined primarily by $J_{nn}$ and $K_p$, then the discrepancy between theory and experiment is of the order of 10%. To compare the exchange integrals with experiment, we can estimate the coefficient ($A$) of the exponential function in the harmonic approximation.[2] In order of magnitude,

$$A \approx z n_e a\left(\frac{M\hbar}{\pi}\right)^{1/2} \omega_D^{3/2}, \tag{4}$$

where $z$ is the number of equivalent tunneling paths from $X_1$ to $X_2$, and $n_e$ is the number of atoms that are exchanged. Figure 1b shows the exchange integrals as functions of the molar volume. All three exchange integrals are comparable in magnitude, confirming that many-body exchange must be incorporated in the Heisenberg Hamiltonian. Curve 4 corresponds to an analysis of data for the $^3$He magnetic pressure according to the Heisenberg model with only nearest neighbors being taken into account. If many-particle exchange processes are taken into account in the analysis of the experimental data, then curve 4 will drop to a lower level.

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