Nuclear Spin-Ordering Due to Correlated Atomic Motion in bcc $^3$He

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We propose a new way to treat nuclear magnetism of solid $^3$He. We argue that the magnetic interaction arises indirectly as a consequence of correlated zero-point motion of the ions. This motion lowers the energy of the ground state, and results in a coherent state of oscillating electric dipoles. Distortion of the electronic wavefunctions leads to hyperfine magnetic interactions with the nuclear spin. Our model describes both the phonon spectra and the nuclear magnetic ordering of bcc $^3$He using a single parameter, the dipolar interaction energy $E_0$. The model yields correctly both the $u2d2$ symmetry of the ordered phase and the volume dependence of the magnetic interaction.

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1. INTRODUCTION

The spin-ordered phase of bcc $^3$He presents a difficult challenge to accurate theoretical description. The main problem is to explain why the transition temperature of $10^{-3}$K is two orders of magnitude larger than the nuclear dipolar interaction $\sim 10^{-5}$K. We propose that magnetic ordering is due to correlations in the zero point atomic motion. In this respect it is similar to the current Multiple-Spin Exchange (MSE) model although the zero-point atomic motion in our model however does not involve the exchange of atoms. This model has conceptual problems that arise from the fact that in order to fit experimental data, many exchange cycles involving a large number of atoms are needed. In fact, it is not sure that this expansion converges, i.e. may not be the spin-ordering mechanism. Additionally,
an overall consistent description of the experimental data has still not been achieved. It is therefore of interest to consider the possibility of another approach.

The zero-point correlations we consider can be described as zero-point electric dipoles and lower the energy of the ground state. This coherent state of oscillating electric dipoles modifies the transverse T1(110) phonon spectrum. We show here that this zero-point motion produces an oscillating magnetic polarization of the electronic cloud which interacts with the nuclear spin. This hyper-fine type interaction has the right order of magnitude to be related to the spin-ordering transition, and leads naturally to the distinct u2d2 antiferromagnetic phase.

2. COHERENT DIPOLES AND TRANSVERSE PHONONS

At temperatures which are high compared to the magnetic interactions (T ≫ 1mK), the local zero-point motion in bcc 3He can be treated in the same way as in 4He. We begin by observing that the crystal potential is highly anisotropic in the bcc phase, being especially wide and anharmonic along the major axes (100,010,001). This means that the atomic wavefunctions will be particularly extended in these directions, resulting in dynamic-correlations, to reduce the overlap energy. The current treatment using variational wavefunctions incorporates these correlations using a Jastrow-type function in a Self-Consistent Harmonic (SCH) calculation. This gives a satisfactory description of the phonon branches, except for the transverse T1(110) phonon, which in reality is much softer. Introducing cubic terms soften the phonon spectra, but now ruins the good agreement with the other branches.

We would like to describe the softening of this transverse phonon as resulting from the hybridization of the (virtual) harmonic phonon with a (virtual) local mode. The coupling is described as dipolar, where this zero-point dipole moment arises due to correlated atomic motion in the normal axes. The harmonic description of the crystal potential misses the low-lying vibration of the atoms due to the shallow (even double-well) potential in the normal directions, which is the one responsible for the dynamic correlations. We therefore treat them as two independent degrees of freedom. In this hybridization procedure we do rely on the SCH calculation, taken from previous works.

Directional oscillation of the nucleus will break the rotational symmetry of the nuclear position relative to the electronic cloud. Going beyond the Born-Oppenheimer approximation, the energy due to relative nuclear-electronic fluctuations is $\Delta E \simeq (m/M)E_{sp} \simeq 10K$, where m is the elec-
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![Diagram showing two degenerate 'antiferroelectric' dipole arrangements in the ground-state of the bcc phase.](image)

Fig. 1. The two degenerate 'antiferroelectric' dipole arrangement in the ground-state of the bcc phase. The arrows show the instantaneous direction of the electronic dipoles.

...tron mass, \(M\) the nuclear mass, and \(E_{sp}\) the excitation energy of the He. This is of the order of the energy involved in the zero-point mixing of the \(s\) and \(p\) electronic levels: \(|\psi\rangle \approx |s\rangle + \lambda |p\rangle\), i.e. \(\lambda \sim \sqrt{\Delta E/E_{sp}} \sim 10^{-2}\). Since the nuclear motions are perfectly correlated this polarization results in zero-point dipolar interactions. The dipolar interaction energy \(E_0\), of the order of \(\Delta E\), is the energy associated with the zero-point correlated oscillations of the atoms along the major axes.

The ground-state of the crystal in which the zero-point motion of the atoms is correlated may be described as a global state of quantum resonance between the two degenerate configurations shown in Fig.1, each of which minimizes the dipolar interaction energy.

Due to the lower symmetry of the dipolar array (Fig.1) compared to the crystal symmetry, a coupling between the harmonic phonons and the dipolar modulation exists only along the \((110)\) direction. At the edge of the Brillouin zone this transverse phonon has the energy \(E_0\). In addition there appears a localized excitation of energy \(2E_0\), which is involved in mass diffusion and contributes to the specific-heat, i.e. a quantum analogue of a point defect. There is good agreement of our approach with the experimental data for the \(T_1(110)\) spectrum for bcc \(^4\)He. This approach has the advantage over current techniques of explicitly describing the main quantum effect of the anisotropic and anharmonic potential. It also resolves previous inconsistencies relating to the properties of vacancies.

In bcc \(^3\)He, sound velocity data indicates that the slope of the \(T_1(110)\) phonon is about half of the SCH calculation. We predict that this ratio should indeed be 0.5, and therefore take half the energy of the calculated SCH \(T_1(110)\) phonon at the edge of the Brillouin zone to be the bare dipole-
flip energy $E_0 \simeq 5\text{K}$ at $V=21.5\text{cm}^3/\text{mole}$. According to our model, the energy of the localized mode involved in thermally activated self-diffusion is $2E_0 \simeq 10\text{K}$. This value is in excellent agreement with the activation energy measured by x-ray diffraction, ultrasonics and NMR experiments\cite{13} at $V=21.5\text{cm}^3/\text{mole}$. A similar activation energy is also obtained from the excess specific heat\cite{14} and pressure measurements\cite{15}. We therefore establish the likely occurrence of coherent zero-point dipoles in the ground-state of bcc $^3\text{He}$.

### 3. MAGNETIC INTERACTIONS

We now describe the magnetic interaction arising from our model. The lowest $|p\rangle$ level of the He atom has the electrons in a spin $S = 1$ state due to strong exchange interaction\cite{16} of the order of $0.25\text{eV}$. In addition this level is split into 3 sublevels with $J = L + S = 0, 1, 2$. The splitting, due to spin-orbit coupling\cite{16} is of the order of $\sim 1.5\text{K}$. In the ground state, the $|p\rangle$ electrons will reside in the $^3P_2$ sublevel, with an oscillating magnetic moment $M_e$ of size $\sim \lambda m_e$, where $m_e$ is the magnetic moment of an electron. Because part of the magnetic moment is now in the $|p\rangle$ state, there appears a net uncanceled moment of equal size in the $|s\rangle$ component of the
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In $^3$He the nuclear magnetic spin $I = 1/2$ will interact with the oscillating electronic magnetic moment, mainly due to the contact term of the $|s⟩$ electron at the nucleus. The magnetic interaction is of the hyper-fine type and the energy associated with it, $E_{mag}$, is given by: $E_{mag} = \left\langle -\frac{8\pi}{3} M_e \cdot M_n \delta(r) \right\rangle$, where $M_n$ is the nuclear magnetic moment and the calculation of the matrix element follows. We show below that the maximum value of $E_{mag}/k_B \approx 0.75$ mK (for $V=24$ cm$^3$/mole), much larger than the direct nuclear dipole-dipole interaction, and is of the right magnitude to explain the high transition temperature of nuclear ordering in bcc $^3$He.

In our model, the magnetic energy will change with pressure due to changes of the electronic magnetic polarization $M_e$. The main effect as the solid is compressed, is that the 3 sub-levels ($J = 0, 1, 2$) broaden into partially overlapping bands. The effect of the broadening of the sub-levels with pressure can be approximated using the overlap integral of the three sub-levels: $\langle M_e(V) \rangle \simeq (1 - F(V)) \lambda(V)m_e$, where the overlap integral $F(V)$ is calculated for the spin-orbit levels using a simple band calculation. The level broadening is the Coulomb energy of the $|p⟩$ electrons due to overlapping wavefunctions on neighboring He nuclei.

We find that the overlap factor $1 - F(V)$ is very sensitive to volume, changing from 1 at $V=24$ cm$^3$/mole to $\sim 0.01$ at $V=19$ cm$^3$/mole. One can see that as the volume decreases the broadening of the bands increase, thereby decreasing the net magnetic polarization of the oscillating electronic cloud. The strength of the magnetic interaction should be proportional to the measured Curie-Weiss temperature $\theta$. In Fig.2 we compare the normalized magnetic splitting $2E_{mag}$ with the normalized values of the measured $\theta$. We find that the volume dependence of agrees very well.

We now consider the symmetry of the ordered spin system. The existence of the hyper-fine splitting means that the simple quantum resonance condition on each site is broken. Now, the two antiferroelectric configurations shown in Fig. 1 are not degenerate, with an energy difference of $2E_{mag}$ per site. It is possible to restore the degeneracy of the overall ground state, and hence the quantum resonance condition. The possible arrangements of the nuclear spins that fulfill the resonance requirement on each simple sublattice are those that ensure an equal number of atoms with electronic and nuclear spins aligned (and anti-aligned) in both degenerate configurations of the electronic dipoles. These arrangements preserve the overall time-reversal symmetry of the system at zero field. We therefore end up with an u2d2 arrangements which is the symmetry of the ordered nuclear phase. We point-out that the u2d2 phase results from symmetry considerations, independent of any quantitative parameters (as in MSE).
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4. CONCLUSION

To conclude, our model enables us to describe the nuclear magnetic ordering of bcc $^3$He using a single parameter, which is the thermal activation energy $E_0$. Experimental values of $E_0$ were measured in several experiments. The model describes correctly both the symmetry of the ordered phase and the volume dependence of the magnetic interactions.

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REFERENCES

1. M. Cross and D. Fisher. Rev. Mod. Phys. (1985).
2. M. Cross, Jap. J. of App. Phys. 26 (1987) 1855.
3. M. Roger, J.H. Hetherington and J.M. Delrieu, Rev. Mod. Phys. 55 (1983) 1.
4. W.P. Halperin, F.B. Rasmussen, C.N. Archie and R.C. Richardson, J. Low Temp. Phys. 31 617 (1978).
5. N. Gov and E. Polturak, Phys. Rev. B 60 (1999) 1019.
6. D.S. Greywall and P.A. Busch, Phys. Rev. B 36 (1987) 6853.
7. The reduction in the ground state energy due to this coherent state is of the same order as the energy difference between the hcp and bcc solid phases (N. Gov, to be published in Phys. Rev. B July 2000, cond-mat/9904249).
8. Y. Masuda and H. Suzuki, J. Low Temp. Phys. 75 (1989) 159.
9. H.R. Glyde, Excitations in Liquid and Solid Helium, Oxford Series on Neutron Scattering in Condensed Matter, 1994.
10. D.S. Greywall, Phys. Rev. A 3 (1971) 2106.
11. T.R. Kohler, Phys. Rev. Lett. 18 (1967) 654.
12. I.B. Bersuker and V.Z. Polinger, ‘Vibronic Interactions in Molecules and Crystals’,1983, Springer Series in Chemical Physics 49.
13. S.M. Heald, D.R. Baer and R.O. Simmons, Phys. Rev. B 30 (1984) 2531.
14. D.S. Greywall, Phys. Rev. B 15 (1977) 2604.
15. I. Iwasa and H. Suzuki, J. Low Temp. Phys. 62 1 (1986).
16. C. Cohen-Tannoudji, B. Diu and F. Laloe, Quantum Mechanics, Vol. II, Wiley-Interscience (1977).
17. N.W. Ashcroft and N.D. Mermin, Solid State Physics, Saunders College Publishing, 1976.
18. D.D. Osheroff, M.C. Cross and D.S. Fisher, Phys. Rev. Lett. 44 (1980) 792.
19. A. Benoit, J. Bossy, J. Flouquet and J. Schweizer, J. Phys. Lett. 46 (1985) L923.