Bcc $^4$He as a Coherent Quantum Solid: 
"Super-Solid"?

Nir Gov

Department of Physics, University of Illinois at Urbana-Champaign, 1110 Green St., Urbana 61801, IL, USA

In this work we investigate the quantum nature of bcc $^4$He. We show that it is a solid phase with an Off-Diagonal Long Range Order of coherently oscillating local electric dipole moments. These dipoles arise from the correlated zero-point motion of the atoms in the crystal potential, which oscillate in synchrony so that the dipolar interaction energy is minimized. This coherent state has a three-component complex order parameter. The condensation energy of these dipoles in the bcc phase further stabilizes it over the hcp phase at finite temperatures. This condensation of the dipoles is not a 'super-solid'. We further show that there can be fermionic excitations of this ground-state and predict that they form an optic-like branch in the (110) direction.

PACS numbers: 67.80-s,67.80.Cx,67.80.Mg

1. INTRODUCTION

The bcc phase of $^4$He has a pronounced quantum nature due to the relatively open structure of the lattice. Quantum effects are manifested in strong anharmonicity of some phonon modes and in the large zero-point kinetic energy of the atoms. In a previous paper we have proposed a new physical model for the local atomic zero-point motion in the bcc phase. In this model we assume that there exist in the bcc $^4$He a phase with coherently oscillating and anisotropic local electric dipoles. The ground-state of these coherent dipoles minimizes the dipolar interaction energy between them. We find that bosonic phase fluctuations in the (110) direction reproduce the spectrum of the anomalously soft $T_1(110)$ phonon. Local dipolar flipping results in a new optic-like branch, described using Fermi statistics.
Fig. 1. (a) The coherent zero-point motion executed by the atoms along the normal directions in the crystal potential. The large arrows show the configuration of the oscillating dipoles along one of the normal axes. (b) The coherent oscillations of the ion in the double-well potential distorts the electronic cloud. This creates the oscillating electric dipole moments.

2. GROUND-STATE COHERENT DIPOLES

In our model, we focused on the effects of the local zero-point motion of the atoms inside the potential-well on the nature of the ground-state. Since in the directions normal to the unit cube’s faces (i.e. (100), (010) etc.) the confining potential well of an atom due to its neighboring atoms is very wide with a pronounced double-minimum structure (Fig.1), the dynamic correlations between the atoms along these directions will tend to keep them apart, by virtually exciting the atom to oscillate between the minima of the potential along the (100) direction, correlated with similar zero-point motions of the other atoms (quantum resonance). If we relax the Born-Oppenheimer approximation, and allow some relative motion between the ions and the electrons, we will obtain that the motion of the ions creates an oscillating electric dipole. This oscillating electric dipole moment $\mu$, is equivalent to a mixing of the $s$ and $p$ electronic levels (Fig.1). Since these zero-point dipole moments are correlated between the different atoms, the resulting dipolar interactions are minimized by the configuration of Fig.1a. It is the dipolar interactions that drive the correlated zero-point nuclear oscillation with energy $E_0$. The oscillating state of quantum resonance of each atom is of the form: $|\Psi(t)\rangle = |s\rangle + \lambda e^{iE_0t/\hbar}|p_{x,y,z}\rangle$, where $|s\rangle,|p_{x,y,z}\rangle$ are the electronic ground-state and first excited state, and $\lambda \simeq 0.01$ being the mixing of the levels due to the oscillation of the nucleus. The system oscillates in resonance
Bcc \(^{4}\)He as a Coherent Quantum Solid: ”Super-Solid”?

between the equivalent up/down arrangements of the ground-state dipoles (Fig.1), simultaneously arranged along all three orthogonal axes.

The effective Hamiltonian for the interacting coherent dipoles:

\[ H_{loc} = \sum_{k} (E_{0} + X(k)) \left( b_{k}^{\dagger} b_{k} \right) + \sum_{k} X(k) \left( b_{k}^{\dagger} b_{-k}^{\dagger} + b_{k} b_{-k} \right) \]  

(1)

where \( b_{k}^{\dagger}, b_{k} \) are Bose creation/annihilation operators of the local mode, \( E_{0} \) is the energy of exciting a local dipole out of the correlated ground-state, and \( X(k) \) is:

\[ X(k) = -|\mu|^{2} \sum_{i \neq 0} \frac{3 \cos^{2}(\mu \cdot (r_{0} - r_{i})) - 1}{|r_{0} - r_{i}|^{3}} \exp[2\pi i k \cdot (r_{0} - r_{i})], \]

where \( \mu \) is the oscillating electric dipole moment, perpendicular to the wavevector \( k \) which is along the (110) (or equivalent) direction, where modulations of the array of dipoles shown in Fig.1 correspond to phonons of the lattice.

The Hamiltonian \( H_{loc} \) which describes the effective interaction between localized modes can be diagonalized using the usual Bogoliubov transformation. The new ground-state is a coherent state, where excitations have the energy spectrum:

\[ E(k) = \sqrt{E_{0} (E_{0} + 2X(k))}. \]

The bare energy \( E_{0} \) to flip a local-dipole out of the coherent ground-state is \( E_{0} = -2X(0) \), i.e. twice the ground-state dipolar interaction energy. The empirical value of \( E_{0} \simeq 7K \) taken from NMR data therefore fixes the size of the coherent dipole moment \( \mu \). Since the phonon is a modulation of the relative phases of the atomic motion, it is therefore equivalent to the excitation described by \( E(k) \). The agreement we find by comparing \( E(k) \) with the transverse \( T_{1}(110) \) phonon data taken from inelastic neutron scattering emphasizes the self-consistency of our description. Further comparisons of this model with experimental data can be found in.

3. ODLRO AND CONDENSATION

The coherent ground-state defines a global phase and breaks the global gauge symmetry of a well-defined occupation number of local dipoles. In the limit \( k \to 0 \) we find that the occupation number of the local-modes diverges as \( 1/k \), signaling macroscopic Bose-Einstein condensation in the zero-momentum state: \( \langle n_{k} \rangle \to_{k \to 0} \frac{1}{2} \frac{E_{0}}{E(k)} = \frac{E_{0} / 2}{E_{c} k^{2} c}, \) where \( c \) is the sound velocity of the \( T_{1}(110) \) phonon which is the natural excitation of the dipolar array. This is just the order of divergency in the occupation number which is typical of an interacting Bose system where it can be related to the occupation of the zero-momentum state, i.e., the condensate fraction \( n_{0}/n \):

\[ \langle n_{k \to 0} \rangle = \frac{m_{0}}{n} \frac{m c}{2\hbar k}, \]

where \( m \) is the boson mass. Since we describe local excitations whose total number is not a conserved quantity, their condensate
fraction can not be simply defined. Nevertheless the coherent dipoles do correspond to the behavior of the $^4\text{He}$ atoms so comparing the residues of the divergent part in these two expressions we find an effective condensate fraction:

$$\frac{n_0}{n} = \frac{E_0/2}{mc^2} \simeq \frac{3.5}{19} = 35 \pm 8\%,$$

where $c$ is $\sim 130 - 160\text{m/sec}$. We note that the condensation of the local modes is only in the directions (110). This means that the limited (vanishing in the $T=0$ case) phase space volume will decrease the overall condensate fraction. At the bcc temperatures ($\sim 1.4\text{K}$) the one dimensional chains in the (110) directions are thermally broadened so that the condensation is now over finite volume sections of phase space. We can estimate this effect, and find a condensate fraction: $n_0/n \sim 0.5\%$.

Similar condensation of local dipoles along all three orthogonal axes of local zero-point motion means that there are three independent phases at each lattice site. The order-parameter can be described as a vector of three complex functions of independent phase: $\Phi(r) = (|\mu| e^{i\theta_x(r)}, |\mu| e^{i\theta_y(r)}, |\mu| e^{i\theta_z(r)})$, where $|\mu|$ is the size of the coherent dipole moment. The bcc $^4\text{He}$ is therefore a system having both Diagonal Long Range Order (DLRO) of the solid lattice and Off-Diagonal Long Range Order (ODLRO) of the local dipoles. It is not a 'super-solid' in that it does not contain both a superfluid and a solid, but is more similar to the superconductors which have a DLRO of the atoms in the lattice and ODLRO of the superconducting electrons. Recent experiments on the behavior of implanted metallic atoms (Cs) in solid $^4\text{He}$ reveal strong coherence effects in the bcc phase, in accordance with our expectation of a coherent state of dipoles.

The condensation energy of the dipoles lowers the energy of the ground-state of the bcc phase and further stabilizes it with respect to the hcp phase:

$$\Delta E = \sum_k \frac{E(k) - (E_0 + X(k))}{2} < 0,$$

which is negative since $X(k) < 0$. We can estimate this sum at the finite temperature of the bcc phase: $\Delta E \simeq -2\text{mK}$ per atom. This result is in agreement with the experimentally interpolated energy difference between the bcc and hcp phases of solid $^4\text{He}$, which is of the order of a few mK per atom.

### 4. Fermionic Excitations

In addition to the fluctuations of the phase of the coherent dipole ground-state (i.e. $T_1$ (110) phonons), there can be a 'dipole-flip' mode, where a dipole is in anti-phase (phase difference of $\pi$) relative to the ground-state configuration of the dipoles. As we mentioned before, this is just the definition of the bare local-mode energy $E_0$. Such an excitation is naturally treated as a fermion since a flipped dipole is antisymmetric with respect to its ground-state configuration, that is with respect to the global phase of
Bcc $^4$He as a Coherent Quantum Solid: ”Super-Solid”? 

Fig. 2. The spectrum of the fermionic optic-like mode ($E_f(k)$, solid line) compared with the experimentally measured phonons in the (110) direction [9].

$\Phi(\mathbf{r})$. The effective Hamiltonian describing such a fermion should contain a term describing the creation and annihilation of pairs of fermions from the ground-state by a $T_1(110)$ phonon. In addition there is a term that describes the excitation energy of the bare fermionic localized-mode, i.e. $E_0$. The effective Hamiltonian that we therefore propose is

$$H_D = \sum_k E(k) \left( c^\dagger_{-k} c^\dagger_k + c_k c_{-k} \right) - E_0 \sum_k c^\dagger_k c_k$$  \hspace{1cm} (2)$$

where $c^\dagger_k, c_k$ are the creation/annihilation operators of the flipped dipoles. In the absence of the second term we have just the Bose ground-state rewritten in terms of fermion pairs. We linearize and solve the equations of motion that follow from (2), similar to the BCS method [4] with the resulting energy spectrum: $E_f(k) = \sqrt{E_0^2 + E(k)^2}$. In Fig.2 we plot the energy spectrum $E_f(k)$ compared with the other phonon modes in the (110) direction. No other optic-like phonons are expected in the bcc lattice. These predictions await high-resolution neutron, Raman and x-ray scattering experiments to be compared with. Note also that these excitations are confined to the (110) directions so despite their relatively low energy, they do not contribute significantly to the specific heat of the solid.
5. CONCLUSION

We have identified a three component complex order parameter and Bose-Einstein condensation in the bcc solid phase, though not a 'super-solid'. There can be further manifestations of the ODLRO of the dipoles in the bcc phase which we have not explored yet, such as macroscopic topological defects in this complex order-parameter. We further obtain that a locally flipped-dipole will behave as a fermion, with an optic-like branch in the (110) direction.

ACKNOWLEDGMENTS

I thank Emil Polturak for useful discussions and encouragement. This work was supported by the Fulbright Foreign Scholarship grant, the Center for Advanced Studies and NSF grant no. PHY-98-00978.

REFERENCES

1. H.R. Glyde, *Excitations in Liquid and Solid Helium*, Oxford Series on Neutron Scattering in Condensed Matter, (1994).
2. N. Gov and E. Polturak, *Phys. Rev. B* 60, 1019 (1999).
3. L.H. Nosanow *Phys. Rev. B* 146, 120 (1966).
4. J.J. Hopfield, *Phys. Rev.* 112 (1958) 1555.
5. P.W. Anderson, *Concepts In Solids*, p.132-148, (1963).
6. W. R. Heller and A. Marcus, *Phys. Rev.* 84, 809(1951).
7. I. Schuster, Y. Swirsky, E.J. Schmidt, E. Polturak, and S.G. Lipson, *Europhys. Lett.* 33, 623 (1996); A. R. Allen, M. G. Richards, and J. Schratter, *Jour. Low Temp. Phys.* 47, 289(1982).
8. J. Gavoret and P. Nozieres, *Analas of Physics*, 28, 349 (1964).
9. V. J. Minkiewicz, T. A. Kitchens, G. Shirane, and E. B. Osgood, *Phys. Rev. A* 8, 1513 (1973).
10. P.E. Sokol and W.M. Snow, *J. Low Temp. Phys.* 101, 881 (1995).
11. V. J. Minkiewicz, T. A. Kitchens, F. P. Lipschultz, R. Nathans, and G. Shirane, *Phys. Rev.* 174, 267 (1968).
12. A.F. Andreev, *Prog. in Low T. Phys. VIII* (1982); A. Leggett, *Phys. Rev. Lett.* 25 1543 (1970); A. Widom and D.P.Locke, *J. Low Temp. Phys.* 23, 335 (1976); M.J. Bijlsma and H.T.C. Stoof, *Phys. Rev. B* 56, 14631 (1997).
13. W. Kohn and D. Sherrington, *Rev. Mod. Phys.* 42, 1 (1970).
14. S. Kanorsky, S. Lang, T. Eichler, K. Winkler and A. Weis, *Phys. Rev. Lett.* 81, 401 (1998).
15. D.O. Edwards and S. Balibar, *Phys. Rev. B* 39, 4083 (1989).
16. C. Kittel, *Quantum Theory of Solids*, John Wiley & Sons, Inc., (1970).