Bcc $^4$He as a Coherent Quantum Solid

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In this work we investigate implications of the quantum nature of bcc $^4$He. We show that it is a unique solid phase with both a lattice structure and an Off-Diagonal Long Range Order of coherently oscillating local electric dipole moments. These dipoles arise from the local motion of the atoms in the crystal potential well, and oscillate in synchrony to reduce the dipolar interaction energy. The dipolar ground-state is therefore found to be a coherent state with a well defined global phase and a three-component complex order parameter. The condensation energy of the dipoles in the bcc phase stabilizes it over the hcp phase at finite temperatures. We further show that there can be fermionic excitations of this ground-state and predict that they form an optical-like branch in the (110) direction. A comparison with 'super-solid' models is also discussed.

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I. 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 atom. It is this large kinetic energy which is thought to help stabilize the bcc phase over the hcp phase. In this paper we highlight this nature of the bcc phase by proposing a new physical model for the local atomic motion. For the sake of clarity we have reproduced here some of the arguments and calculations already given in\textsuperscript{1}. We propose that in the bcc $^4$He phase the local excitations of the atoms in their potential wells, result in oscillating local electric dipoles. The ground-state of these dipoles has the dipoles oscillating in synchrony, thereby reducing the dipolar interaction energy between them. Solving a mean-field Hamiltonian describing these dipoles we find that Bosonic phase fluctuations in the (110) direction reproduce the spectrum of the T$_1$(110) phonon.

In the following we further explore the nature of the coherent ground-state of the local-modes in the bcc $^4$He. We show that the bcc $^4$He is a unique phase having both Diagonal Long Range Order (DLRO) of the solid lattice and Off-Diagonal Long Range Order (ODLRO) of the local dipoles. There is therefore a complex three-component order parameter which describes the coherently oscillating dipoles in each of the three orthogonal directions in the lattice. In the ground-state the local dipoles form a Bose-Einstein condensate in the zero momentum state, and we are able to estimate the ground-state energy reduction due to this condensation. This estimate compares favourably with experimental results and consequently we claim that this condensation energy stabilizes the bcc phase over the hcp phase. In the hcp phase we expect no coherence or condensation due to the highly isotropic lattice and the geometric frustration of the hexagonal symmetry. We also comment about the relation of this work to previous work about the 'super-solid' concept in quantum solids.

Additionally, we predict a high-energy optical-like mode which has fermionic statistics. This excitation is confined to the (110) direction and involves a local 'flipping' of a dipole with respect to the ground-state. This makes the dipole become anti-symmetric (a $\pi$ phase difference) with respect to the global phase of the complex order parameter and acquire Fermi-Dirac statistics. We also give analytic expressions for the scattering intensity of both the Bose and Fermi excitations along the (110) direction. These predictions remain to be compared with future experimental data.

II. GROUND-STATE COHERENCE AND BOSE EXCITATIONS.

The usual treatment of the ground-state and energy of bcc $^4$He employs variational wavefunctions that aim to account for the short-range correlations between the atoms. These correlations arise mainly due to the hard-core repulsion between the atoms. The atoms have a high zero-point kinetic energy which is given quite accurately by treating them as independent particles held in place by the potential of the neighboring (static) atoms. This type of calculation is the ”particle-in-cell” approximation which gives surprisingly good agreement with measured thermodynamic properties of the solid phase.

We want to focus here on the effects of the local motion of the atoms inside this potential well on the nature of the ground-state. In this approach we would like to isolate the lowest energy excited state of the atom inside its potential well, and treat it as a local excitation of the lattice. This local excited state consists of a local oscillatory motion of the atom along a particular direction and produces an oscillating electric dipole, similarly to that...
of the usual Van-der Waals interaction. However unlike the case of the Van-der Waals interaction, in which the dipolar fluctuations are random, we show that in the bcc solid there are local dipoles which are correlated and a new ground-state of lower energy is created.

The potential well of an atom in the bcc lattice due to the standard helium pair-potential \( v(r) \), provided one can take the other atoms as stationary, can be mapped along any direction in the lattice. We find that in the directions normal to the unit cube’s faces (i.e. (100), (010) etc.) the confining potential well is very wide with a pronounced double-minimum structure (Fig.1). Solving the one-dimensional Schrodinger’s equation for a \(^4\)He atom in this potential, we get a first excited level with energy \( 10K \), and a wavefunction describing atomic motion with an amplitude of \( \sim 1\AA \) (in the (100) direction (Fig.1)). Based on the above calculation, we shall assume that the atoms have a local-mode that is highly directional along one of the directions equivalent to (100). Local atomic motion along the other directions is assumed to be severely restricted due to the higher excitation energies (Fig.1). Experimental evidence for the existence of such a "local mode" comes from NMR measurements which find an activation energy of \( 7.17K \), and we propose to identify this local-mode with the highly directional motion of the atoms in the normal directions.

Using this identification we can now estimate the size of the local electric dipole moment that can be created by this local and highly directional atomic motion. As the atom moves this instantaneous local electric-dipole is created due to the electronic cloud and the ion being slightly displaced relative to each other. The electric dipole moment due to mixing of the lowest \( |s\rangle \) and \( |p\rangle \) electronic-levels of the \(^4\)He atom, is given from perturbation theory as

\[
\psi = |s\rangle + \lambda |p\rangle \Rightarrow E_0 \simeq \langle \psi | E \psi \rangle - \langle s | E | s \rangle \simeq \lambda^2 \langle p | E | p \rangle
\]

\[
\lambda^2 \simeq 7 \times 10^{-18} \text{ eV} \quad \lambda \simeq 0.0168 \tag{1}
\]

where \( |s\rangle \) and \( |p\rangle \) stand for the ground-state and first excited-state of the \(^4\)He atom, \( \lambda \) is the mixing coefficient and \( \langle p | E | p \rangle \simeq 2.46 \cdot 10^{-18} \text{ eV} \) is the excitation energy of the first atomic excited-state. This small estimated mixing gives the magnitude of the induced dipole moment as

\[
|\mu| = e \langle \psi | x | \psi \rangle \simeq 2e \lambda \langle s | x | p \rangle \simeq e \cdot 0.03 \AA \tag{2}
\]

where \( \langle s | x | p \rangle \simeq 0.9 \AA \). The estimation of the mixing \( \lambda \) and the dipole-moment \( |\mu| \) serves to set an upper bound on the magnitude of this effect, since we assumed that the entire excitation energy \( E_0 \) is converted to a local electric dipole.

It is possible to show that the lowest energy of a correlated dipolar array in the bcc lattice preserves the symmetry of the bcc unit cell along one of the symmetry axes. In such a case it can be easily shown that there will be no contribution to the dipolar interaction energy from oscillating dipole moments which are orthogonal, and the instantaneous dipolar interaction energy for each of the three orthogonal directions, is given by

\[
E_{dipole} = - |\mu|^2 \sum_{i \neq 0} \left[ \frac{3 \cos^2 (\mu \cdot |r_0 - r_i|) - 1}{|r_0 - r_i|^3} \right] \tag{3}
\]

where the sum is over all the atoms in the lattice, \( r_i \) being the instantaneous coordinate of the \( i \)-th atom. For oscillating dipoles with random phases, the average instantaneous interaction energy \( \overline{E_{dipole}} \) summed over the lattice would be zero. However, the energy of the dipoles can be lowered by correlating the phases of the oscillating atoms. Since the direction of the local dipole shows the instantaneous direction of the motion or displacement, a state where all the dipoles point in the same direction is just a uniform motion or translation of the entire lattice. We therefore have to look for symmetric arrangements with respect to the number of up/down dipoles, such as is shown in Fig.2. This is the lowest energy "antiferroelectric" configuration with the periodicity of the bcc unit cell. We have shown this arrangement for individual dipoles oriented along the (001) direction, but they are similarly arranged for dipoles along the two other orthogonal axes. The sum in (3) for such a configuration with a unit dipole is given in Fig.2. Thus, the ground-state in our picture has the atoms executing this correlated local oscillation along the three orthogonal directions.

We therefore have, in addition to the usual (isotropic) Van-der Waals interaction, highly directional (anisotropic) electric dipoles that become correlated so that they oscillate in synchrony. This is a state of quantum resonance where the system oscillates between two equivalent up/down arrangements of the ground-state of the dipoles (Fig.2). The total interaction between the atoms is now given as the usual second-order (\( \propto 1/r^6 \)) Van-der Waals contribution that is the result of local-dipoles which have random relative phases, and an additional long-range (first-order, \( \propto 1/r^3 \)) dipolar interaction from the correlated part. Dipolar interactions that decay as \( 1/r^3 \) occur for perfectly correlated oscillating dipoles, such as a single electric dipole and its image in an adjacent conducting plate. The coherently oscillating nearest-neighbor dipoles therefore behave as perfect images of each other (Fig.2), and oscillate with the same global phase.

The correlated oscillating dipoles do not have an average static dipole moment, so this is not the case of an antiferroelectric structural phase transition. The array shown in Fig.2 is simultaneously arranged along the other two orthogonal axes. Along each direction the ground-state is given as a coherent-state of these local dipoles, i.e. has a well-defined phase and an ill-defined occupation number.

We shall treat the dynamics of the correlated dipole array as independent of the other degrees of freedom of the lattice. This assumption needs justification since there can be phonon modes that will modulate
the atomic motion, thereby coupling with the oscillating dipolar array. The oscillatory atomic motion induced by the phonons will modulate the relative phases of the dipoles. Let us look at the ground state of the dipoles, taking for example dipolar oscillations oriented along the (001) direction (Fig.2). We now need to consider only phonons which will modulate the local motion responsible for the oscillating dipoles in this direction. In the bcc structure, only 3 phonons fulfill this condition: L(001), T(100) and T_1(110). Let us calculate the energy of the dipolar array when modulated along these 3 directions. For a modulation along some direction \( k \), the dipolar interaction energy is given by:

\[
X(k) = -|\mu|^2 \sum_{i \neq 0} \left[ 3 \cos^2 \left( \frac{\mu \cdot (r_0 - r_i)}{|r_0 - r_i|^3} \right) - 1 \right] \exp \left[ 2\pi i k \cdot (r_0 - r_i) \right] \tag{4}
\]

At \( k = 0 \) the interaction matrix \( X(k) \) is just the dipolar energy \([3]\).

In Fig.3 we plot the value of \( X(k) \), the energy of the dipolar array modulated by the relevant phonons: L(001), T(100), and T_1(110), for dipole moment \(|\mu| = 1\). We see that for a modulation by L(001) and T(100) the periodicity of \( X(k) \) is over a full unit-cell, that is twice the periodicity of these phonons. Since symmetric functions of periodicities \( \pi/a \) and \( 2\pi/a \) are orthogonal, so are the eigenfunctions of these particular phonons and dipole-excitations. The dipole array cannot therefore be excited by any of these two phonons. For the modulation produced by the T_1(110) mode, the periodicity of \( X(k) \) is the same as that of the T_1(110) phonon, which can therefore couple to the dipole array. We conlude therefore that the coupling of the local modes to the lattice excitations is limited to a single phonon mode, justifying our assumption that the local modes can be treated separately to a good approximation. We shall now calculate the dispersion relation of such an excitation by a mean-field solution of an effective Hamiltonian. It turns out that the only phonon mode of the bcc lattice that can couple with the dipolar array is in fact the natural excitation of the dipolar array in the (110) direction. Thus, the only elementary (Bose) excitations of the dipole array would be in the (110) direction, in the form of the T_1(110) phonon. The description of this phonon is therefore taken into account by our treatment of the dynamics of the dipolar array, and will appear as a solution of the mean-field treatment. This means that our assumption of an effective decoupling between the dipolar and other degrees of freedom is justified.

The Hamiltonian treatment of interacting local excitations was developed originally by Hopfield \([4]\) for the problem of excitons in a dielectric material. The local excitations are treated as bosons using the standard Holstein-Primakoff procedure, and the effective Hamiltonian describing their behavior is \([4]\):

\[
H_{loc} = \sum_k \left( E_0 + X(k) \right) \left( b_k^\dagger b_k + \frac{1}{2} \right) + \sum_k X(k) \left( b_k^\dagger b_{-k}^\dagger + b_k b_{-k} \right) \tag{5}
\]

where \( b_k^\dagger, b_k \) are Bose creation/annihilation operators of the local mode, \( X(k) \) is given in \([4]\) and \( E_0 \) is the energy of exciting a local dipole out of the correlated ground-state.

The Hamiltonian \( H_{loc} \) \([4]\) which describes the effective interaction between localized modes can be diagonalized using the Bogoliubov transformation \( \beta_k = u(k)b_k + v(k)b_{-k}^\dagger \). The two functions \( u(k) \) and \( v(k) \) are given by:

\[
u^2(k) = \frac{1}{2} \left( \frac{E_0 + X(k)}{E(k)} + 1 \right), \quad v^2(k) = \frac{1}{2} \left( \frac{E_0 + X(k)}{E(k)} - 1 \right) \tag{6}
\]

The result of solving by mean-field the effective Hamiltonian for the correlated dipolar array \([4]\) is a coherent ground-state given by:

\[
|\Psi_0\rangle = \prod_k \exp \left( \frac{\nu_k}{u_k} b_k^\dagger b_{-k}^\dagger \right) |\text{vac}\rangle \tag{7}
\]

and the energy spectrum is

\[
E(k) = \sqrt{E_0 \left( E_0 + 2X(k) \right)} \tag{8}
\]

In Fig.2 we see that the ground-state arrangement has the dipoles arranged in alternating planes in the (110) direction. As we have shown the only naturally occurring Bose excitations of this dipolar field are along this direction and \( X(k) \) is the dipolar interaction matrix element for \( k \) in the (110) direction (Fig.3). In order to calculate the energy spectrum we now need to fix the size of the coherent dipole moment \(|\mu|\). According to our definition of the local mode the energy cost of flipping the direction of a single dipole out of the ordered ground state is defined to be \( E_0 \). This is equivalent to having \( 2|X(k = 0)| = E_0 \), which is the condition to have a gapless mode at \( k \to 0 \) \([4]\). Using this condition, the experimental value of \( E_0 = 7k_B T \) determines the size of the coherent dipole moment as: \(|\mu| \approx e \cdot 0.01 \text{Å} \). This value is indeed smaller than our previous estimation, which served as an upper bound on the size of the oscillating dipole moment \([4]\).

As we have proposed, the phase modulation in the (110) direction of the transverse atomic motion in the lattice, with energy \( E(k) \) \([6]\) should coincide with the T_1(110) phonon. In Fig.4 we compare the experimental values of T_1(110) taken from neutron scattering data with the calculated \( E(k) \), and we find that the agreement is excellent for all \( k \). From \([6]\) and Fig.3 we see that at the edge of the Brillouin zone the energy \( E(k) \) of the phonon should be just the bare energy of the local mode, \( E_0 \), since \( X(\sqrt{2}\pi/a) = 0 \). We also have that...
at \( k = \sqrt{2\pi/a} \) the dipoles have changed between the two configurations illustrated in Fig.3, which are the two possible configurations with alternating dipoles arranged on adjacent planes with the periodicity of the bcc unit cell.

Since the empirical value of \( E_0 \) that we used was taken from NMR data, the agreement we find with the phonon data taken from inelastic neutron scattering, emphasizes the self-consistency of our description. We stress that the value of \( E_0 \) and the lattice vectors are the only empirical inputs used in the calculation, with the functional behavior completely given by the lattice structure and the dipolar interactions.

**III. OFF-DIAGONAL-LONG-RANGE-ORDER AND CONDENSATION.**

We have found from the mean-field solution at zero temperature that the ground-state of the bcc phase contains a coherent-state of oscillating local-dipoles. Since our method predicts the excitation spectrum of the \( T_1(110) \) phonon with very good accuracy, we expect it to be valid at the finite temperatures for which the bcc phase exists. We therefore expect that the basic nature of the bcc phase will be well described by our results, although the quantitative values may change due to the finite temperature. The coherent ground-state defines a global phase and breaks the 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

\[
\langle n_k \rangle = v^2(k) = \frac{1}{2} \left( \frac{E_0 + X(k)}{E(k)} - 1 \right) \to k \to 0 = \frac{E_0}{2} E(k) = \frac{E_0/2}{2\hbar c k} \tag{9}
\]

where \( c \) is the sound velocity of the \( T_1(110) \) phonon which is the natural excitation of the dipolar array. This is identical to the result for a Weakly Interacting Bose Gas (WIBG) problem solved by Bogoliubov where the divergence is related to the occupation of the zero-momentum state, i.e., the condensate fraction \( n_0/n \)

\[
\langle n_k \rangle_{WIBG} = v^2(k) \to k \to 0 = \frac{n_0 kmc}{n 2\hbar k} \tag{10}
\]

where in the WIBG case we have \( c \) as the \( k \to 0 \) sound velocity, and \( \varepsilon_k = \hbar^2 k^2/2m \) is the free particle energy. By comparing (9) with (10) we find that in the bcc case the role of the condensate fraction, the WIBG order-parameter, is taken by the parameter \( E_0 \), which is just 2\( |X(0)| \). This can be seen directly from the form of the ground-state wavefunction where the pair-occupation is given by:

\[
\langle b_k b_{-k} \rangle = 2u(k)v(k) = \frac{X(k)}{E(k)} \tag{11}
\]

Equating the divergent part in (9) and (10) we can define an effective condensate fraction

\[
\frac{n_0}{n} = \frac{E_0/2}{mc^2} \approx 3.5 \frac{35 \pm 8%}{10} \tag{12}
\]

where we used for the velocity of sound \( c \) the values from our calculation \( \approx 130m/\text{sec} \) and from elastic constants \( \approx 160m/\text{sec} \). It must be remembered that the mass \( m \) in (12) is not necessarily the mass of a bare \( ^4\text{He} \) atom since we are now dealing with condensation of local dipoles. Comparing with the condensate fraction at zero temperature in superfluid \( ^4\text{He} \) which is \( \approx 10\% \), we find that it is lower than the condensate fraction of the local modes in the bcc phase. We again mention that our result is for \( T=0 \) which can be depleted at finite temperature.

It is clear that it is a non-zero coherent dipole moment \( \mu \) that produces a dipolar interaction matrix \( X(k) \) which in turn implies finite pair occupation (1) and a coherent ground-state. This is just the condition for the Bose-Einstein condensation of the dipoles in the bcc ground-state. We therefore have a broken gauge symmetry and a complex order-parameter in the form of the pair-occupation (11). This function can be complex since the conditions on \( u(k) \) and \( v(k) \) allow for a relative complex phase between them, just as in the WIBG case. A similar condensation of local dipoles in all three orthogonal axes of local motion means that there are three independent phases at each lattice site, since orthogonal dipoles do not interact. The order-parameter in our case can therefore be described as a vector of three complex functions of independent magnitude and phase:

\[
\Phi(r) = \begin{pmatrix} |\mu_x| e^{i\theta_x(r)} \\ |\mu_y| e^{i\theta_y(r)} \\ |\mu_z| e^{i\theta_z(r)} \end{pmatrix} \tag{13}
\]

If the cubic symmetry is not broken by external stresses, the magnitude of the coherent dipole moment in the three orthogonal directions should be the same: \( |\mu_x| = |\mu_y| = |\mu_z| \). In the ground-state the phases are spatially uniform, while the excited state is described through a periodic phase oscillation, i.e. the \( T_1(110) \) phonon. The order parameter (13) is to be contrasted with the order parameter of superfluid \( ^4\text{He} \), which also exhibits ODLO and which has a single complex component.

In the hcp phase we do not expect the dipoles to order in a coherent state since the hexagonal geometry frustrates antiferroelectric-type configurations. Also the nearly isotropic potential of the hcp lattice does not allow the highly directional dipole moments as in the bcc case. Indeed there is good agreement between experiments and the harmonic calculation of the phonons in the hcp phase, indicating no strong quantum corrections, as in the bcc phase.

The bcc \( ^4\text{He} \) is therefore a unique crystallographic phase 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. This system is also distinct from the case of Bose-Einstein condensation of a phonon mode which results in a static deformation of the lattice and a structural phase-transition.

Bose-Einstein condensation of local defects (vacancies) was previously considered for solid $^4$He, and is similar to our treatment. The main difference is that in our case the physical picture of the condensed local modes is not a local distortion of the lattice like a vacancy, and that the condensation is unique to the bcc phase. The estimate in these works is that in the ground-state ($T=0$) the density of vacancies is $\sim 10^{-7}$ per site. In contrast we expect in the bcc phase a sizable fraction (10-30%) of condensed local-modes per site.

The fact that the region of existence of the proposed supersolid phase in the phase diagram of solid $^4$He should closely coincide with the region occupied by the bcc phase was shown in [4]. In this work it was further shown that in the supersolid there should be a second-sound-like mode, which is an oscillation in the density of the defects. In our description of the bcc phase this suggests the possibility of an oscillation in the amplitude of the order-parameter $\Phi(r)$, that is in the amplitude of the coherently oscillating dipole moment. This is in contrast to the $T_1(110)$ phonon mode which is an oscillation in the phase of the order-parameter. This mode may be produced by modulating the density of the atoms so that the local excitations energies change and with them the amplitude of local motion and local electric dipole moment. Unfortunately we do not expect such a mode to have measurable consequences which are different from the effects produced by usual longitudinal phonons.

In concluding this section, we would like to mention the recent experiments of the behaviour of implanted metallic ions (Cs) in solid $^4$He. These experiments are designed to look for evidence of time-reversal symmetry breaking which is equivalent to having a static electric dipole moment. In our description of the bcc phase we do not find a static but a coherent-dynamic electric dipole moment. We point out that in these experiments a marked difference between the hcp and bcc phases has been found. In the bcc phase the electronic-spin relaxation of the Cs atom is extremely slow and this effect could be a result of the coherence and long-range order of the dipolar fields. The coherently oscillating $^4$He electrons in the bcc phase will produce a very uniform electromagnetic interaction with the electronic spin in the Cs atom. By comparison, in the hcp phase the spin polarization is extremely short lived, indicating a more random field environment. This result is in accord with our expectation that the coherent dipoles are unique to the bcc phase. Similar experiments in the future may allow a probe that will show directly the coherently oscillating dipoles in the bcc ground-state.

In these experiments the hyperfine transition in the Cs atom was also measured. The energy shift of this transition is sensitive to the shape of the confining cavity of the Cs atom inside $^4$He lattice. The width of the transition is a measure of the fluctuations in this cavity size, and the data show a much smaller spread in the bcc compared with the hcp phase. Uncorrelated atomic motions of the $^4$He atoms will increase the spread in instantaneous cavity sizes due mainly to breathing-like motion of the cavity walls (Fig.5). This behavior is what we expect for the hcp case. The correlated atomic motion in the bcc phase should result in a more constant cavity shape (Fig.5) and a narrow signal, which is indeed measured.

IV. GROUND-STATE ENERGY AND THE STABILITY OF THE BCC PHASE

The question of the relative stability of the different crystal structures in solid He has been a long standing one. The necessity for some non-Van-der-Waals interactions has been previously proposed to explain the occurrence of fcc over hcp structure in the heavier rare-gas solids. The bcc phase is usually found to be more stable than the close-packed hcp phase due to the large zero-point energy in the He solids. The correlations between the dipoles in the ground-state that we have proposed, lowers the energy of the ground-state of the bcc phase and further stabilizes it with respect to the hcp phase. The reduction in ground-state energy achieved by the coherent state of the dipoles along one of the three orthogonal directions, is given by

$$\Delta E = \sum_k \frac{E(k) - (E_0 + X(k))}{2E_0} < 0 \quad (14)$$

which is negative since $X(k) < 0$ and $E(k) < (E_0 + X(k))$.

Since the energy reduction integrand (14) is non-zero only in the $(110)$ directions it will give a small contribution to the three dimensional phase-space integration. At zero temperature the summation in (14) will be confined to one dimensional sections along the $(110)$ direction, so that the contribution will be zero. At the bcc temperatures ($\sim 1.4K$) the one dimensional chains in the $(110)$ directions are broadened so that the summation in (14) is now over finite volume sections of phase space. We can estimate the maximum width of the conical section in $k$-space as the momentum which corresponds to a $T_1(110)$ phonon with energy $k_BT$, that is $\sim 0.13\text{A}^{-1}$. The numerical integration of (14) over such volume sections gives an energy reduction of $\Delta E \sim -2\text{mK per atom}$. This result is in agreement with the experimentally interpolated energy difference between the bcc and hcp phases of solid $^4$He, which is of the order of a few mK per atom.

This reduction is less than 0.1 percent of the potential and kinetic energies of the solid, and is therefore
very hard to calculate accurately theoretically\footnote{4}. What is shown in the usual calculations is that the correlations between the motions of the atoms are essential in lowering the energy of the bcc phase, compared with the hcp phase. Since part of the correlations in the atomic motion is described by our coherent dipole model, we expect the condensation energy of the dipoles (14) to be important in determining the stability of the bcc phase. At finite temperature the stabilization of the bcc phase compared to the hcp phase is usually attributed to the lower zero-point energy due to the lower T\(_1\) (110) phonon energy\footnote{5}. This is just the phonon which is softened by the long-range dipolar interactions that we have described, indicating again the importance of the coherent dipoles to the stabilization of the bcc phase of solid \(^3\)He. Our procedure may provide a good estimate of the small change in energy at the structural phase transition, by isolating the degree-of-freedom which is most affected by the transition, i.e. the correlated atomic motion along the directions normal to the unit cell faces.

The picture we propose is that the dipole condensation mechanism of the bcc phase competes with the lower potential energy of the hcp phase due to its higher coordination number. If the hcp phase has a large enough volume (through thermal expansion or introduction of \(^3\)He impurities) its potential energy is increased until a critical point is reached where the bcc phase has a lower total energy due to the dipolar-condensation energy reduction\footnote{6}, which is absent in the hcp phase\footnote{4}. At this critical point the structural phase transition occurs.

By comparison, solid \(^3\)He has a stable bcc phase due to the larger kinetic energy of this lighter isotope. This increased zero-point energy causes the less dense bcc phase to have a lower ground-state energy than the hcp phase even at T=0. Since the \(^3\)He atoms are fermions with a spin 1/2 nucleus, the oscillating electric dipoles are not in resonance and can not be treated as bosons\footnote{4}. We therefore do not expect a coherent state of the atomic motion as in the bcc \(^4\)He, and bcc \(^3\)He is stable due to its large zero-point kinetic energy alone. On the other hand, at low enough temperatures where the bcc \(^3\)He becomes an antiferromagnet, there could be correlations involving both the nuclear spin and electric dipole degrees of freedom.

V. 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 localized ‘flip’ of a dipole so that it is in anti-phase (phase difference of \(\pi\)) relative to the rest of the dipoles, in the ground-state configuration. Such an excitation is naturally treated as a Fermion since such a flipped dipole is antisymmetric with respect to the other dipoles, that is with respect to the global phase \(\theta\) (13) in one (or more) of the orthogonal directions of local motion \((x, y, z)\). The flipped dipole is no longer a dipolar image of the nearest-neighbor dipoles but an anti-image, and will be treated with Fermi-Dirac statistics.

An anti-phase localized-mode (a fermion) is not part of the correlated ground-state, but nevertheless will feel the effect of the Bose excitations (T\(_1\) (110) phonons) of the dipolar array as they interact with it. The effective Hamiltonian describing such a fermion should therefore contain a term describing the creation and annihilation of pairs of fermions from the ground-state by a phonon (Boson). This is an off-diagonal term that describes the fluctuation caused by a T\(_1\) (110) phonon of energy \(E(k)\): it changes a fermion ‘particle’ into a ‘hole’ and vice versa. The terms ‘particle’ and ‘hole’ are with respect to the ground-state which has occupation of pairs of localized-modes (i.e. not an ‘empty’ vacuum).

In addition there should be a term that describes the excitation energy of the bare fermionic localized-mode, that is \(E_0\). This is just the energy to ‘flip’ a dipole from the ground-state so that all it’s interactions with the other dipoles of the ground-state change sign, i.e. \(-2X(0) = E_0\).

The many-body effective Hamiltonian that we therefore propose is

\[
H_D = \sum_k E(k) \left( c_k^\dagger c_{-k} + c_k c_{-k} \right) - \sum_k V_k \left( c_k^\dagger c_k c_{-k}^\dagger c_{-k} \right)
\]

(15)

where \(c_k^\dagger, c_k\) are the creation and annihilation operators of the anti-phase (Fermionic) localized-mode. The first term in (13) is the ‘kinetic’ term due to the phonon-roton branch, where the localized-modes are created/annihilated in pairs. The energy \(E(k)\) is the energy of the T\(_1\) (110) phonon excitation (8). In addition there is a finite ‘potential’ energy if there is a finite density of unpaired fermions, which is \(E_0\). In the absence of the second term we have just the Bose ground-state written in terms of fermionic pairs.

We linearize the equations of motion that follow from (16), similar to the BCS method\footnote{24},

\[
i\hbar \dot{c}_k = -E(k)c_{-k}^\dagger + \Lambda_k c_k \quad i\hbar \dot{c}_k^\dagger = -E(k)c_k - \Lambda_k^* c_{-k}^\dagger
\]

(16)

where we used the Fermi anti-commutation relations:

\[
\{c_k, c_{k'}^\dagger\} = 1 \quad \{c_k, c_{-k'}\} = 0, \text{ and we define}
\]

\[
\Lambda_k = \Lambda_k^* \equiv E_0 = \sum_k V_k \left( c_k^\dagger c_k \right), \quad \sum_k \left( c_k^\dagger c_k \right) = 1, V_k = E_0
\]

\[
\Lambda_k \equiv 1
\]

(17)

From (17) we see that the symbol \(E_0\) will now indicate a finite density of fermions.

The equations of motion have the following eigenvalues:
$$\begin{vmatrix} E_f(k) - E_0 & -E(k) \\ -E(k) & E_f(k) + E_0 \end{vmatrix} = 0 \Rightarrow E_f(k) = \sqrt{E_0^2 + E(k)^2}$$

(VII. CONCLUSION)

In this work we have investigated the nature of the quantum correlations in the bcc phase of solid $^4$He. We identified a three component complex order parameter and Bose-Einstein condensation in this phase, though not a ‘super-solid’ i.e. no superfluid component. 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 the complex order-parameter. The order-parameter or condensate-fraction can also serve as an extra thermodynamic variable, and this opens the possibility of more complicated internal dynamics in the bcc solid, such as the phenomenon of second sound in superfluid $^4$He.

We predict that a local excitation of a dipole out of the coherent ground-state will behave as a Fermion, and we calculate its energy spectrum. We find it to behave as an optical-like branch in the (110) direction. Finally we calculate the scattering intensity as a function of wavevector $k$ for both the Bose ($T_{11}(110)$ phonon) and Fermi (new optical-like branch) excitations. All these predictions await high-resolution neutron and Raman scattering experiments to be compared with.

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VIII. APPENDIX A: COMPARISON OF BOSE EXCITATIONS WITH KLEIN-GORDON HAMILTONIAN.

We would like to point out that the Hamiltonian describing the localized dipoles is similar to the Klein-Gordon (KG) Hamiltonian for a single spinless boson, written in its first-order form.

Klein-Gordon:

\[ H_{KG} = \varepsilon_k (\sigma_z + i \sigma_y) + mc^2 \sigma_z + e\Phi \hat{\imath} \]

(24)

Localized dipoles:

\[ H_{loc} = X(k) (\sigma_z + i \sigma_y) + E_0 \sigma_z \]

(25)

where \( \sigma_i \) are the Pauli matrices, \( \varepsilon_k = \vec{p}^2/2m \), \( e \) is the electric charge, \( \Phi \) is the electrostatic potential, \( m \) is the KG-particle’s mass and \( c \) is the velocity of light.

We have written the dipolar hamiltonian (25) in the basis of a two component wavefunction

\[ \Psi_{loc} = \begin{pmatrix} c^\dagger_k \\ c_{-k} \end{pmatrix} \]

(26)

In this representation we see that exciting a local dipole out of the ground-state configuration (\( c^\dagger_k \)) has bare energy \( E_0 \) while destroying an excited dipole has minus this energy. There is a freedom of choice weather to define the positive excitation to be a flipping of an up dipole to a down dipole or vice versa. The sign of the energy of the dipolar bosons therefore represents this freedom which corresponds to two equivalent dipolar configurations with a \( \pi \) phase difference.

The two-component wavefunction of the KG Hamiltonian is:

\[ \Psi_{KG} = \begin{pmatrix} \varphi \\ \chi \end{pmatrix} \]

\[ \varphi = \frac{1}{2} \left( \psi + \frac{i\hbar}{mc^2} \psi^0 \right), \chi = \frac{1}{2} \left( \psi - \frac{i\hbar}{mc^2} \psi^0 \right) \]

(27)

where \( \psi \) is the original wavefunction of the second-order KG equation, and \( \psi^0 = (\frac{\Phi}{\hbar} + \frac{i}{\hbar} \Phi) \psi \). The Hamiltonians (24)(25) are the similar except that the KG density is not normalized to 1 but to \( \langle \rho \rangle = E/mc^2 \), describing the relativistic increase in the density with velocity. By comparing the two Hamiltonians (24)(25) we identify that: \( E_0 \leftrightarrow mc^2 \), \( X(k) \leftrightarrow \varepsilon_k \), which gives the equivalence of the two hamiltonians.

The peculiarities of the KG equation appear when there is a potential \( V = e\Phi \) (the Klein paradox for example). The equation for the momentum of the KG particle:

\[ 2me^2 \varepsilon_k = \hbar^2 c^2 k^2 = (E(k) - V)^2 - (mc^2)^2 \]

\[ \Rightarrow k = \sqrt{(E(k) - V)^2 - (mc^2)^2} \]

(28)

becomes the equation for \( X(k) \) in the dipolar case:

\[ X(k) = \frac{(E(k) - V)^2 - (E(k)_0)^2}{2E_0} \]

(29)

We see from (29) that there is a region of energies where the interaction parameter \( X(k) \) is positive and a region where it is negative. We saw above that the condensation of the dipoles in the bcc phase is characterized by a negative \( X(k) \) which also gives a gapless excitation spectrum at \( k \rightarrow 0 \). The excitations with \( E(k) > V + E_0, E(k) < V - E_0 \) are therefore not contributing to the coherent long-range order. A fermionic excitation is a local destruction of the coherent order, and indeed costs at least \( E_0 \) (for the free case with \( V = 0 \)) to create (19).

In the case of the KG equation the sign of the energy indicates the charge of the particle/antiparticle, which have opposite charges. Charge conjugation therefore interchanges between the two. What is the meaning of the different signs of the energy of the dipolar boson excitations in the bcc case? From our definition of the second-quantized description of the dipoles, the meaning of the sign of the energy is that the field of resonating localized-dipoles can have two global configurations shifted by \( \pi \) (Fig.2). These two configurations are identical with respect to the magnitude of the energy spectrum, but in each the operation of spin flip changes from up→down to down→up. We can therefore identify two "charges" for the bcc to distinguish between the two shifted phases. Further we find that as in the KG case the operation of charge conjugation (which reverses the signs of the dipoles) moves us between the two solutions.

IX. APPENDIX B: SYMMETRY BREAKING OF THE FERMIONIC EXCITATIONS.

We see from (20) that when there is no fermion present (i.e. if we put \( E_0 = 0 \) in (17)) the ground state has equal numbers of fermions and holes. The symmetry between particles and holes is broken by the free fermion quasiparticle (or quasihole), and the sign of the symmetry-breaking parameter \( E_0 \) determines which of the two kinds is present. The hole/particle are with respect to the equilibrium occupation by pairs of fermions in the ground-state.

A single flipped dipole described as the fermionic excitation, breaks the symmetry between the number of up/down dipoles and creates a residual globally oscillating dipole moment. The parity \( P \) symmetry with respect to reflection along the axis of the global dipole (let us choose to be \( z \)) is broken. The charge \( C \) symmetry is also broken since the direction of the global dipole is flipped under charge conjugation. The time reversal symmetry \( T \) is unbroken since the oscillating global dipole does not define a unique time direction. We therefore have that the global \( CPT \) symmetry is preserved, as is the \( CP \) and
As shown in (31), the time component of the momentum-symmetry that is broken by choosing a non-zero parameter in the 2D Dirac equations-of-motion (32). The broken parity two-dimensions is the time-reversal symmetry (TRS). The symmetry-breaking parameter in (15) is the sign given to $E$. The symmetry-breaking parameter in (15) using:

$$H = \sum_{k} c_k \sigma^z c_{-k} - \frac{1}{2} \sum_{k} V_k (\sigma^z_k + 1)$$  \hspace{1cm} (30)

while the 2D Dirac particle is described by

$$i\hbar \frac{\partial}{\partial t} \left( \begin{array}{c} \varphi \\ \chi \end{array} \right) = c(\vec{\sigma} \cdot \vec{p}) \left( \begin{array}{c} \varphi \\ \chi \end{array} \right) + mc^2 \sigma_z \left( \begin{array}{c} \varphi \\ \chi \end{array} \right)$$  \hspace{1cm} (31)

By assuming momentum $\vec{p}$ in the $\hat{x}$-direction only we write

$$i\hbar \frac{\partial}{\partial t} \left( \begin{array}{c} \varphi \\ \chi \end{array} \right) = c \left( \begin{array}{cc} 0 & -i\hbar \partial_x \\ 0 & 0 \end{array} \right) \left( \begin{array}{c} \varphi \\ \chi \end{array} \right) + mc^2 \left( \begin{array}{cc} 1 & 0 \\ 0 & -1 \end{array} \right) \left( \begin{array}{c} \varphi \\ \chi \end{array} \right)$$  \hspace{1cm} (32)

where $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$, $\sigma_i$ the Pauli matrices, and $\varphi, \chi$ are the particle/antiparticle scalar wavefunctions. There is now complete analogy between (31) and (32). The symmetry-breaking parameter $E_0$ is identical to the $mc^2$ parameter in the 2D Dirac equations-of-motion [22]. The symmetry that is broken by choosing a non-zero $mc^2$ in two-dimensions is the time-reversal symmetry (TRS). As shown in (31), the time component of the momentum-energy vector in two dimensions is taken by the $z$ axis. The broken parity $P$ in the $z$-axis for the fermionic excitation of the bcc phase is here replaced by the TRS breaking of the heavy two-dimensional Dirac particle.

Similar to the Anderson transformation of the BCS problem to a magnetic Hamiltonian we can transform (33) using:

$$n_k = c_k^\dagger c_k \quad c_k^\dagger c_{-k} = \sigma^z_k / 2 \quad c_k c_{-k} = \sigma^+_k / 2$$

$$\Rightarrow n_k n_{-k} = \frac{1}{2} (\sigma^z_k + 1) \quad c_k^\dagger c_{-k} + c_k c_{-k} = \sigma^z_k$$  \hspace{1cm} (33)

where the $\sigma^z_k$ are Pauli spin-1/2 operators. The basis is such that an up-spin in the $\hat{z}$-direction represents an empty pair, while a down-spin represents an occupied pair.

The resulting Hamiltonian is:

$$H_{mag} = \sum \varepsilon_k \sigma^z_k - \frac{1}{2} \sum V_k (\sigma^z_k + 1)$$  \hspace{1cm} (34)

This Hamiltonian describes a fictitious magnetic field acting on the spin $\vec{\sigma}$:

$$\vec{B} = \varepsilon_k \hat{x} - \frac{1}{2} E_0 \hat{z}$$  \hspace{1cm} (35)

where we replaced the potential energy with the constant $V_k = E_0$ [17]. The magnetic field (35) can be compared with the BCS result [10]

$$\vec{B}_{BCS} = \varepsilon_k \hat{x} + \frac{1}{2} V \sum (\sigma^x_k + \sigma^y_k)$$  \hspace{1cm} (36)

The alignment of the spins in the ground-state is shown for the two Hamiltonians in Fig.8. In the BCS problem the sign of the symmetry-breaking field in $V$ (34) has to be positive so that it induces ferro-magnetic interaction between the fictitious spins, otherwise there will not be any rotation of the spins across the Fermi-energy. Only when the spins rotate do they go through the point where the spin is entirely in the $xy$-plane. At this point the state has no defined occupation number but a well defined phase, while on both sides of the domain-wall there is well defined occupation and no phase. This superconducting-phase at the fermi energy is just the angle of the fictitious spin in the $xy$-plane, and there is broken U(1) symmetry.

In the Dirac case the symmetry-breaking field $E_0$ in (33) is a constant external field. It can have both signs, which control the direction along the $z$-axis that the rotated spin has in the middle of the domain-wall. This spin describes weather a particle or a hole is occupied, while away from the $k = 0$ point the spins are in the $xy$-plane, with a well defined phase. The symmetry that is broken is therefore the binary $Z(1) (\pm)$ symmetry.

In comparison with the BCS problem we see that in the fermionic excitation in the bcc the symmetry-breaking parameter is a finite density of unpaired fermions: $\langle c_k^\dagger c_k \rangle \neq 0$ [17]. The ground-state without unpaired fermions is a 'vacuum' of pairs of particle-holes in equal numbers. In the BCS problem the symmetry-breaking parameter is a finite pair-density: $\langle c_k^\dagger c_{-k} \rangle \neq 0$. The ground-state in the absence of electron pairing is just a finite density of electrons below the fermi-energy and zero above. In this respect the two problems are 'complementary'.
FIG. 1. The potential-well of an atom in bcc $^4$He along different directions. The energy difference $E_0$ between the lowest two energy levels (dashed lines) are: (111)- 59.5K, (110)- 27.6K, (100)- 10.6K.

FIG. 2. The coherent dipole arrangement in the ground-state of the bcc phase, oscillating between these two configurations. Dipoles with same phase have same shade. The sum of the dipole-dipole interaction (Eq.3) for a unit dipole-moment is: -0.08 (Å$^{-3}$).
FIG. 3. The calculated interaction matrix $X(k)$ (Eq.4) as a function of the wavevector $k$, for the three phonon modes that could affect a dipolar array. The dipole moment has been normalized to give a gapless mode: $X(k=0) = -E_0/2$. The unit-cell dimensions (Å): $a = 4.12/2$, $a' = a\sqrt{2}$. Also shown are the two arrangements of the dipoles in the extreme points along the oscillation in the (110) direction.

FIG. 4. The experimental data [13] (solid circles) for the $T_1(110)$ phonon compared with the calculation (Eq.8) (solid line). Also shown is the energy of the bare local-mode $E_0 = 7K$.

FIG. 5. Schematic cavity shapes for a Cs atom (grey circle) inside $^4$He solid (empty circles with arrows). (a) Correlated atomic motion in bcc: constant cavity shape, (b) uncorrelated atomic motion in hcp: randomly fluctuating cavity shape.
FIG. 6. The spectrum of the fermionic optic-like mode (Eq.18, solid line) compared with the experimentally measured phonons in the (110) direction [13].

FIG. 7. The calculated intensity of the inelastic neutron scattering by the $T_1(110)$ phonon (Eq.22, solid line) and by the new fermionic mode (Eq.23, dashed line).

FIG. 8. Arrangement of the spins in the magnetic-analogue-Hamiltonian. BCS: (a) without pairing, (b) with pairing. Dirac: (c) without unpaired fermion, (d) with unpaired fermion.