Velocity dependent interactions and a new sum rule in bcc He

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Recent neutron scattering experiments [PRL, 88, p. 195301 (2002)] on solid $^4$He, discovered a new optic-like mode in the bcc phase. This excitation was predicted by a recently proposed model that describes the correlated atomic zero-point motion in bcc Helium in terms of dynamic electric dipole moments. Modulations of the relative phase of these dipoles between different atoms describes the anomalously soft $T_1(110)$ phonon and two new optic-like modes, one of which was recently found in the neutron scattering experiments. In this work we show that the correlated dipolar interactions can be written as a velocity dependent interaction. This then results in a modified f-sum rule for the $T_1(110)$ phonon, in good agreement with the recent experimental data.

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The phonons in a quantum solid, such as solid He, show various quantum effects arising from the unusually large zero-point motion of the atoms. The strong quantum pressure in solid He lowers the density and introduces large anharmonic terms in the interatomic potential. In bcc He the potential even has a double-well feature. The Debye-Waller factor $d(k)$ is unusually small due to the large spread in the atomic positions. In addition there have been measurements of apparent oscillations of the Debye-Waller factor as a function of the scattering wavevector $k$. These oscillations seemed to be confined to some modes, while absent in the transverse $T_1(110)$ mode. The explanation of these unusual oscillations was given in terms of interference effects between the single and multi-phonon excitations, whereby intensity is transferred between them. The mixing of the phonon modes is attributed to the anharmonicity of the interatomic potential. From symmetry considerations these types of calculations predict that the oscillatory contribution to $d(k)$ vanishes both at the Brillouin zone center and boundary (z.b.).

Recently there has been a new neutron scattering experiment in solid $^4$He. The main surprise in this experiment was the discovery of a new, gapped excitation branch in bcc $^4$He. This excitation was found at the energy predicted by a new model of the dynamic effects of the zero-point motion on the phonon spectra. Usually a bcc lattice is not expected to support optic-like phonons, so the measurement of this new excitation branch provides support for the basic idea of this model to describe some of the correlations in the atomic zero-point motion in bcc He. The main point is that there is an anharmonic, low energy mode of atomic motion along the major axes of the bcc lattice, which is not described by the harmonic approximation. In bcc He this mode has energy $E_0 \sim 3 - 10K$ and is the only free parameter in our model, which we take from empirical data. Additionally, the low mass of the He atom allows relatively large breaking of the Born-Oppenheimer approximation which results in motion-induced electric dipoles, due to relative displacement of the nuclei with respect to the electronic cloud. When these dipoles are uncorrelated they introduce a negligible addition to the usual Van-der Waals interaction. By contrast, correlated and highly directional atomic motion results in dipolar interactions between these dipoles, which is of the order of the energy of the local atomic motion $E_0$. There can therefore arise a state of quantum resonance between these interacting electric dipoles and the atomic motion. A resonant state between the atomic motion and electric dipolar interactions lowers the ground-state energy of the system. In the ground-state of the bcc phase we found that the dipolar interaction is minimized when the dipoles have the phase relation shown in Fig.1.

The dipolar interactions between the motion-correlated dipoles is

$$E_{dip} = \sum_{i \neq 0} \mu_0 \cdot \mu_i \left[ \frac{3 (\hat{\mu} \cdot \hat{r}_{i0})^2 - 1}{|\hat{r}_{i0}|^3} \right]$$

where the electric dipole moment in bcc He turns out to be $\mu_i \approx e \cdot 0.01$, and $\hat{r}_{i0} = \hat{r}_0 - \hat{r}_i$ and the index $i$ runs over all atoms. The resonance condition means that $E_0$ is the frequency of both the oscillating dipoles and the atomic motion, i.e. $E_{dip} = -E_0/2 \approx -3K$ in bcc $^4$He. Note that the dipole moments are dynamic, and the expectation value of the dipole moment of the
crystal and of each atom is zero, as required by time reversal symmetry. It is the matrix element of correlated dipoles ($\mu_0 \mu_i$) which is non-zero, and appears in (1).

The modulations of the relative phases of the dipoles on adjacent atoms coincide with only one phonon mode, due to the lower symmetry of the dipolar ground-state compared with the bcc lattice (Fig. 1). This mode turns out to be the anomalously soft $T_1(110)$ phonon, which agrees very well with the experimental data [2]. In addition our model predicts two new modes which are gapped (optic-like) [3, 5]. One of these modes was recently observed in neutron scattering experiments [3]. Since the dipolar interactions arise from correlated atomic motion, they can be naturally written as a modulation of the correlated dipolar array [2]. This mode [4, 5] has been found by neutron scattering [4].

Our model therefore gives a good description of some effects of the anharmonic potential in bcc He, as they are compared with the bcc lattice (Fig. 1) [2]. This mode agrees very well with the experimental data [2].

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The space representation of the dipolar interaction energy (2) between the zero-point dipoles in the ground-state can now be written in terms of the local momentum operators

$$X = \sum_{i,j,i \neq j} \vec{\mu}_j \cdot \vec{\mu}_i \left[ \frac{3(\vec{\mu} \cdot \vec{r}_{ij})^2 - 1}{|\vec{r}_{ij}|^3} \right]$$

$$= \left( \frac{e}{m \omega_0} \right)^2 \sum_{i,j,i \neq j} \left[ \frac{3(\vec{\mu} \cdot \vec{r}_{ij})^2 - 1}{|\vec{r}_{ij}|^3} \right] \tilde{P}_j \tilde{P}_i$$

(4)

where $\omega_0 = E_0/\hbar$ is the resonance frequency of atomic and dipolar vibration, and the $\tilde{}$ symbol is used for operators.

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justified by the excellent agreement between our calculation of the and the $T_1(110)$ spectrum experimental data.

The new sum rule we find for the $T_1(110)$ phonon is essentially the usual f-sum rule multiplied by the function $2|X(k)|/E_0$. We find that at $k \to 0$, we recover the usual f-sum rule, since $|X(k \to 0)| \to E_0/2$. While the usual f-sum rule follows from particle conservation, the number of excited dipoles out of the correlated ground-state is not strictly conserved. In particular, when the function $X(k)$ goes to zero at the Brillouin zone center. Additionally, the multi-phonon interference term calculated previously for interference terms allows in stray scattering from other phonons with similar energy (at the z.b. the $T_1(110)$ branch meets other phonon branches). At the z.b. with the smaller momentum ($k \simeq 1.85 \text{Å}^{-1}$) there is higher scattering intensity then at the higher momentum z.b. ($k \to 2.85 \text{Å}^{-1}$), as expected for stray scattering from other phonons. Future high resolution data at the zone boundaries is needed to resolve this question.

In this work we presented a modified f-sum rule for the anomalously soft $T_1(110)$ phonon mode of bcc He. We find an apparently oscillating Debye-Waller factor and a complete vanishing of the first moment at the Brillouin zone boundary. The new f-sum rule is qualitatively different from the standard response of a harmonic phonon. This unusual result is due to correlations in the atomic zero-point motion, which can be written in terms of a velocity-dependent interaction. The calculated sum rule is then compared with recent neutron scattering data, which does show some of the predicted features. Combined with the recent observation of a new excitation mode, the sum rule data presented here provides extra support for the proposed model of correlated zero-point electric dipoles in the ground-state of bcc He.

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FIG. 1: The two degenerate 'antiferroelectric' dipole arrangements in the ground-state of the bcc phase. The system is in quantum resonance between these two configurations with resonance frequency $\omega_0 = E_0/\hbar$. The arrows show the instantaneous direction of the dipoles along one of the major axes. Similar dipolar arrays exist along the orthogonal major axes.

FIG. 2: The dipolar interaction matrix element $X(k)$ (Eq. 2) in the bcc phase of $^4$He, along the (110) direction.

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FIG. 3: The ratio $M_1(k)/((k \cdot e(k))^2d(k)^2)$ given by Eq.6 (dashed line), compared with the experimental data for the $T_1(110)$ phonon: crosses [3] and stars [6]. The usual form for a harmonic phonon is given here by the horizontal dotted line.