New sound mode in superfluid He-4 film adsorbed on atomically flat substrate

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Abstract. New sound mode in superfluid He-4 layer adsorbed on graphite surface is theoretically proposed. A dislocation dynamics, which arises in a solid bilayer underlying the superfluid is explicitly considered. The result is compared with recent QCM experiment on Grafoil.

1. Introduction
Graphite surface provides an atomically flat area over a few hundred Å being free from lattice irregularities and impurities [1]. He-4 films of thickness more than two atomic layers adsorbed on the surface undergo superfluid transition with the first two layers solidified. Recently, Hosomi et. al. have observed a slipping behavior between the solid layers; The second atomic layer of solid (2ALS) has a motion relative to the first atomic layer of solid (1ALS), while 1ALS sticks to the graphite surface and its dynamics can be neglected [2]. The purpose of this paper is to show a possibility of a novel sound mode in the superfluid He-4 layer coupled with the motion of 2ALS.

2. Model
As is known, atomically thin films of superfluid He-4 has third sound propagation for ordinary substrates such as glass and Mylar sheet. A normal component of the fluid should stick to the substrate because of the thinness of the films, hence only a superfluid component oscillates by the van der Waals potential of the substrate which acts as a restoring force. In the present case, we anticipate that the normal component also has a dynamics along with the mobile component of 2ALS. We assume that the latter component is edge dislocations of a density \(\rho_d(x,t)\), although easily extendable to the other mass carrier such as interstitials, domain walls and vacancies. For simplicity, we consider a line shape for the dislocation parallel to \(y\)-axis on the glide plane, and moves to \(x\)-direction. \(z\) is the direction normal to the graphite surface. See Figure 1.

'Two fluid' description for the present system consists of two conservation laws

\[
\frac{\partial \rho}{\partial t} = - \nabla \cdot \left( \rho_s \vec{u}_s + \rho_n \vec{u}_n \right),
\]

and two equations of motion:

\[
\frac{\partial (\rho \vec{S}_t)}{\partial t} = - \nabla \cdot \left( \rho \vec{S}_t \vec{v}_n \right),
\]
Figure 1. Configuration of He-4 layers and axes. Circles are schematic cross sections of edge dislocations accommodating in the second atomic layer of solid (2ALS).

\[ \frac{\partial \mathbf{v}_s}{\partial t} = -\nabla \bar{\mu} - f \hat{z} - F \hat{x}, \quad (3) \]

\[ \frac{\partial}{\partial t} (\rho_s \mathbf{v}_s + \tilde{\rho}_n \mathbf{v}_n) = -\nabla P - \rho_\ell f \hat{z} - \tilde{\rho} F \hat{x}, \quad (4) \]

where all the friction terms are neglected and we put \( \rho_s \) and \( \rho_n \) respectively as normal and superfluid mass densities. For abbreviation, we put \( \tilde{\rho}_n \equiv \rho_n + \rho_d \), \( \tilde{\rho} \equiv \rho_s + \rho_n + \rho_d \) and \( \rho_\ell \equiv \rho_s + \rho_n \). The dislocations are always combined with the normal fluid component and drift at velocity \( \mathbf{v}_n \). We consider a case where the dislocations carry negligible entropy compared with what the normal fluid component does, and take \( \bar{S}_\ell \) as the entropy per gram of the fluid. \( \bar{\mu} \) is the Gibbs free energy per gram of the fluid and relates \( \nabla \bar{\mu} = -\bar{S}_\ell \nabla T + 1/\rho_\ell \nabla P \). The system is subject to a pressure gradient \( \nabla P \), the van der Waals potential \( f(z) \) and acceleration \( F \) which can be imposed by external oscillation (such as torsional oscillation and QCM). We note that the dislocations move only within the plane, so \( f(z) \) acts only on the fluid, whereas \( F \) accelerates both the fluid and the dislocations. Also, we assume that the dislocations are not accelerated by \( \nabla P \). In fact, each dislocation has a lattice distortion only locally in an atomic length scale \( w \) [3] in the solid bilayer and can move freely unless the separation between them is shorter than \( w \) [4]. The free dislocation motion causes local in-plane deformation in 2ALS which does not couple to \( \nabla P \). This assumption is hence not appropriate for the case when \( \nabla P \) is significantly large: If the liquid pressure becomes (temporarily) large in some area so as to donate dislocations dense enough to overlap with each other, the dislocations should be accelerated through their repulsive interaction. Such a solid-liquid conversion will appear when \( F \) is great due to the large amplitude of external oscillation.

The coupled equations (1)-(4) lead to an in-phase and an out-of-phase motion between the superfluid component and the normal fluid - dislocation complex, as expected. The present system is thus similar to the case of aerogel filled with superfluid He-4; first sound like mode and forth sound like mode are realized respectively for the in-phase and the out-of-phase motions between the superfluid and the normal fluid component bound to the aerogel matrix [5]. There are, however, two differences: First, the present system is two dimensional and the sound realized in a limit \( \rho_d \rightarrow \infty \) is third sound, not forth sound. Since the superfluid density is kept constant during third sound propagation, we shall take in the present system the total fluid density \( \rho_\ell \) as a constant and relate the pressure to the out-of-plane displacement \( \xi(x, t) \) of the fluid by

\[ P(x, z, t) = \rho_\ell \int_z^{d_2 + d + \xi} dz f(z) + P_g, \tag{5} \]

where \( d_2 \) and \( d \) are the average thicknesses of 2ALS and the fluid layer respectively, and \( P_g \) the pressure of environmental gas. We set \( z = 0 \) on the glide plane (between 1ALS and 2ALS).
The fluid compression is hence absent on the sound propagation: Unlike the aerogel case, the smallness of thermal expansion coefficient is not important to decouple the in-phase motion from the out-of-phase motion of sound. The second point that is different from the aerogel case is the possibility of fluid-solid conversion. That is, a freezing-melting wave can cooperate in the sound propagation of fluid in a novel way [6]. We will find soon, however, that this conversion does not appear in a linear approximation below.

Linearizing the equations (1)-(4) with respect to \( \vec{v}_s \) and \( \vec{v}_n \), we have

\[
\frac{\partial^2 \tilde{\rho}}{\partial t^2} = \Delta P, \tag{6}
\]

\[
\frac{\partial^2 \bar{S}_\ell}{\partial t^2} = \frac{\rho_s \tilde{\rho}}{\rho_\ell \rho_n} \bar{S}_\ell^2 \Delta T - \frac{\rho_s \rho_d}{\rho_\ell^2 \rho_n} \Delta P. \tag{7}
\]

Taking \( T \) and \( P \) as a function of \( \xi \) and \( \bar{S}_\ell \), instead of \( \rho_\ell \) and \( \bar{S}_\ell \) of the aerogel case, and regarding \( (\partial P/\partial \bar{S}_\ell)_\xi \) is negligibly small, we have two decoupled sound modes. One is a pressure wave where the superfluid component oscillates with the normal fluid component bound to the dislocations in an in-phase way at velocity \( c_i \). The other is a temperature wave where the two components oscillate in an out-of-phase way propagating at velocity \( c_o \);

\[
c_i^2 = \frac{\bar{d}}{a} f(\bar{d}), \tag{8}
\]

\[
c_o^2 = \frac{\rho_s \tilde{\rho}}{\rho_\ell \rho_n} \bar{S}_\ell^2 \frac{T}{C_v}, \tag{9}
\]

where we took an integration over the thickness \( 0 < z < d_2 + d + \xi \) for Eq. (6) and used the relation (5). \( \bar{d} \) is \( d_2 + d \) plus 1ALS’s thickness. \( a > 1 \) is a factor of order of unity caused by the fluid mass enhancement \( \tilde{\rho} > \rho_\ell \) due to the coupling between the normal fluid and the dislocations.

The sound velocity (8) resembles that of third sound (without evaporation-condensation process) except that the total fluid density (with an enhancement factor) cooperates in instead of superfluid density. Therefore this sound survives across superfluid transition like first sound in a bulk He-4 does. The temperature wave is, on the other hand, associated with superfluid transition as implied in (9) similar to second sound velocity in the bulk. Apparently \( c_o << c_i \) especially in the vicinity of \( T_c \).

Putting \( \ell \) as the length scale of the surface of graphite, and set \( \omega_i \equiv c_i/\ell \) and \( \omega_o \equiv c_o/\ell \), we note that there are three regions of probe frequency \( \omega \) to the He-4 film to have different dynamical response: I. \( \omega < \omega_o \), II. \( \omega_o < \omega < \omega_i \) and III. \( \omega_i < \omega \). In the high frequency region III, both modes, especially the in-phase deformation is possible to detect. A mass decoupling of \( \tilde{\rho} \) is thus observed. In the region II, the in-phase deformation is absent in average over the period \( 2\pi/\omega \). So the normal fluid flow dragged by the dislocation should be canceled by the superfluid counterflow to keep \( \xi = 0 \): The mass decoupling is hence \( \rho_d \). In the region I, not only the in-phase, but also the out-of-phase deformation of fluid is hard: The dislocation motion dragging the normal fluid component against superfluid has a restoring force in proportion to \( \rho_s \). Growing \( \rho_s \) to some degree, hence the dislocations are locked and the He-4 film totally sticks to the substrate.

3. Comparison with experiment

Recently, Hosomi et. al. have measured a period shift \( \Delta P \) of an oscillating Grafoil (exfoliated graphite) with He-4 adsorption [6]. Adsorbed He-4 forms a film consists of a solid bilayer and a fluid overlayer of a few atomic thickness on a number of connected graphite platelets. They
used QCM (Quartz Crystal Microbalance) technique with frequency $\omega = 10^7$ Hz and amplitude $A = 10^{0-1} \text{Å}$. In small $A$ cases, they observed Kosterlitz-Thouless type superfluid transition being consistent with a torsion oscillation experiment [7]. For large $A$ cases, on the other hand, say $A > A_c$ where $A_c = 4\text{Å}$ typically, a mass decoupling starts to grow below some temperature $T_S$ slightly higher than superfluid transition temperature $T_c$. The mass decoupling is much larger than the superfluid mass decoupling, and is ascribed to the slipping of 2ALS on 1ALS. As shown in Figure 2, with lowering temperature the mass decoupling monotonously increases, and suddenly drops at temperature $T_D$ which is slightly below $T_c$.

An interesting possibility, that is a fluid-solid conversion (a crystallization wave restricted in a two dimensional space) is suggested for the origin of this unusual sticking [6]. Here we have another possibility: The transition from the region II to I of the last section occurs at $T_D$. In fact, taking $\ell = 100 \text{Å}$ by regarding the graphite platelets are mostly disconnected in Grafoil [8], and putting $c_i = 100 \text{ m/s}$ and $c_o = 10 \text{ m/s}$ which are respectively of usual third sound at $T = 0$ and of the bulk second sound near $T_\lambda$ in order, we have $\omega_i = 10^{10}$ Hz and $\omega_o = 10^9$ Hz. Considering that $c_o$ grows from zero (at $T_c$) to 10 m/s (below $T_c$), then the transition from II to I at some temperature below $T_c$ is plausible. Quantitative comparison should be done in future.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure2}
\caption{Schematic result of experiment [6]. There appear two characteristic temperatures $T_D$ and $T_S$ other than $T_c$ at large amplitudes of QCM oscillation. The superfluid mass decoupling at $T_c$ cannot be recognized in this scale.}
\end{figure}

4. Summary
We have shown a possibility of new sound propagation of He-4 films adsorbed on atomically flat substrate such as graphite surface. Due to the flatness of substrate, the solid bilayer can have a dynamics through the motion of edge dislocation. In the presence of superfluid overlayer, hence in-phase motion and out-of-phase motion is possible between the superfluid component and the normal fluid - dislocation complex. The qualitative relevance to recent QCM experiment is noted.

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