Andreev-Lifshitz Hydrodynamics Applied to an Ordinary Solid under Pressure

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We have applied the Andreev-Lifshitz hydrodynamic theory of supersolids to an ordinary solid. This theory includes an internal pressure $P$, distinct from the applied pressure $P_a$ and the stress tensor $\lambda_{ik}$. Under uniform static $P_a$, we have $\lambda_{ik} = (P - P_a)\delta_{ik}$. The theory also permits vacancy diffusion but treats vacancies as conserved. It gives three sets of propagating elastic modes; it also gives two diffusive modes, one largely of entropy density and one largely of vacancy density (or, more generally, defect density). For the vacancy diffusion mode (or, equivalently, the lattice diffusion mode) the vacancies behave like a fluid within the solid, with the deviations of internal pressure associated with density changes nearly canceling the deviations of stress associated with strain. We briefly consider pressurization experiments in solid $^4$He at low temperatures in light of this lattice diffusion mode, which for small $P_a$ has diffusion constant $D_L \sim P_a^2$. The general principles of the theory – that both volume and strain should be included as thermodynamic variables, with the result that both $P$ and $\lambda_{ik}$ appear – should apply to all solids under pressure, especially near the solid-liquid transition. The lattice diffusion mode provides an additional degree of freedom that may permit surfaces with different surface treatments to generate different responses in the bulk.

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I. INTRODUCTION

Since the late 1960’s there have been theoretical suggestions that solids might display flow behavior similar to what is found in superfluids. For that reason there has been a great deal of interest in solid $^4$He as a candidate supersolid. The first experimental indication of superflow was the appearance of a non-classical moment of inertia (NCRI), first observed by Chan’s group, since confirmed by many other laboratories, and strongly linked to disorder. In addition, the shear modulus shows anomalous behavior, although not enough to explain the NCRI experiments. Non-NCRI superflow has been searched for but not observed. We also note recent experiments that argue against any supersolid signature above approximately 55 mK. Further works casting doubt on supersolidity are a study of bcc $^4$He that shows unusual NCRI behavior at higher temperatures, and a study showing that the NCRI behavior due to plasticity has different properties than due to quenching.

In a recent experiment on a pancake-shaped sample, a capacitance gauge monitored the pressure as a function of temperature $T$. Samples were produced by both the slow-cooling blocked capillary method and by the more rapid quench-cooling method, which gives more disordered samples. In one set of measurements the sample was quench-cooled below 1 K in 144 s, during which time the pressure decreased. This is perhaps an indication that vacancies, formed during the quench, were leaving the sample. For a blocked-capillary sample the temperature was lowered below 500 mK while the pressure was monitored vs $T$. The sample was then annealed at 1.65 K, where the pressure increased, perhaps an indication that vacancies now were entering the sample. A second cooldown yielded, by a reduced $T^2$ term in the pressure, an indication that the sample was less disordered, but that disorder remained. Even at a constant temperature of 19 mK the pressure continued to relax, which is consistent with vacancies equilibrating. The fact that the observed relaxation times do not saturate at the temperatures studied indicates that the temperature is not yet low enough that quantum relaxation processes dominate thermal relaxation processes.

This experiment can perhaps be interpreted under the assumption that the system is not supersolid. We have therefore undertaken a theoretical study of the macroscopic flow properties of a one-component ordinary solid.

Our basis is the theory of Andreev and Lifshitz (AL) for the macroscopic behavior of a supersolid. They included volume $V$ as an extensive variable, in addition to $W_{ik} \equiv V w_{ik}$, where $w_{ik}$ is the non-symmetrized strain. This permited them to continuously go to the superfluid limit as $w_{ik}$ becomes irrelevant. The point of the present work is that on eliminating the superfluid variables, the theory should apply to an ordinary solid. We employ a variation on the notation of Ref. 24, which gives a more explicit derivation of the equations of motion and extends Ref. 1 to include nonlinear terms. Note also the theory of Fleming and Cohen for an ordinary solid, which gives equations with a similar structure, and similar modes, but uses a very different notation (and does not consider an applied pressure $P_a$). Both Ref. 1 and Ref. 24 implicitly assume that uniform vacancy number-changing bulk processes are negligible, and neglect interstitials and impurities. Recently Yoo and Dorsey considered the effect of a lattice diffusion mode on light scattering by a supersolid, but also briefly considering an ordinary solid.

As noted by Martin, Parodi, and Pershan the normal system has eight degrees of freedom, given by two scalar thermodynamic quantities (which can be taken to
be the mass density \( \rho \) and the entropy density \( s \) and two vector quantities: the lattice vector \( u_i \) and the velocity \( v_i \) associated with the momentum density \( g_i = \rho v_i \). (With \( m_i \) the atomic mass and \( n \) the number density of \(^4\)He atoms, we have \( \rho = m_i n \).) As a consequence there are eight normal modes. For a uniform infinite system these modes are three pairs of propagating elastic waves and two diffusive modes, one primarily of the temperature \( T \) and the other primarily of \( \partial_i u_i \). In the absence of lattice defects, for a variation \( \delta u_i \) the relationship

\[
\partial_i (\delta u_i) \approx -\delta \rho / \rho \tag{1}
\]

holds, giving the system one fewer degree of freedom, and thus one fewer mode. One can think of this missing mode, associated with the dynamical violation of \( H \), as being associated with vacancies, as noted in Ref. 28.

The present work obtains the diffusion constant and the physical properties of this diffusive mode, for both zero and non-zero \( P_a \). (\(^4\)He must be under \( P_a \approx 25 \) atmospheres to solidify.) We find that the physical character of the mode is that it involves essentially zero stress deviation, because the fluid-like stress (associated with changes in mass density) nearly cancels the solid-like stress (associated with changes in strain).

Allowing vacancies to move permits mass change without lattice motion.\(^{29}\) This allows one to take the fluid limit of zero crystallinity, and study the evolution of the sound velocity as the system evolves from the perfect solid to perfect liquid. By perfect liquid we mean one with no defects, for a variation \( \delta u \), the relationship

\[
\delta \rho / \rho \approx -\delta T / T \tag{11}
\]

Here \( \lambda_{ik} \) of AL is an elastic tensor density (with units of stress (associated with changes in strain).

Section \( \text{II} \) gives the form of AL supersolid theory when restricted to a normal solid, including the possibility of lattice defects. Although we specifically have vacancies in mind, \(^3\)He impurities could be accounted for if its density were included as an additional thermodynamic variable, which would require extension of the AL theory. Note also the case of (two-component) superionic conductors, which includes certain high-temperature alkali halides, where the larger halide ions remain in a lattice but the lattice of the smaller alkali ions “melts.” Section \( \text{III} \) discusses elasticity and internal pressure for a crystal under static and uniform applied pressure \( P_a \), and calculates internal pressure \( P \) and strain. We find that \( P \sim P_a^2 \), so that for small \( P_a \) the effect of \( P \) is very small; see eq. (6).

For small \( P_a \) the strain is largely included in \( P_a \), as expected, but there is a \( P_a^2 \) correction. Section \( \text{IV} \) derives the normal modes for the ordinary solid. Section \( \text{V} \) considers how such modes can be generated (including the possible effect of different surface treatments), and applies the theory to the pressurization experiments.\(^{22}\) Section \( \text{VI} \) provides a summary and our conclusions. Appendix \( \text{A} \) gives the thermodynamics and dynamics of the AL theory for the supersolid. Appendix \( \text{B} \) calculates some thermodynamic derivatives that appear in the normal modes in terms of \( P_a \).

## II. ANDREEV-LIFSHITZ NORMAL SOLID WITH DEFECTS

We employ the primary quantities energy density \( \epsilon \), lattice displacement \( u_i \), and non-symmetrized strain

\[
w_{ik} = \partial_i u_k. \tag{2}
\]

We consider a normal solid by setting \( \rho_s = 0, \rho_n = \rho, \) \( \vec{v}_n = \vec{v}, \) and eliminating the superfluid equation from the equations for the supersolid (given in Appendix\( \text{A} \)).

### A. Thermodynamics

The appropriate thermodynamic equations are

\[
d\epsilon = T ds + \lambda_{ik} dw_{ik} + \mu d\rho + \vec{v} \cdot \vec{g}, \tag{3}
\]

\[
\epsilon = -P + Ts + \lambda_{ik} w_{ik} + \mu \rho + \vec{v} \cdot \vec{g}, \tag{4}
\]

\[
0 = -dP + s dt + w_{ik} d\lambda_{ik} + \rho d\mu + \vec{v} \cdot \vec{g}. \tag{5}
\]

Here \( \lambda_{ik} \) of AL is an elastic tensor density (with units of stress (associated with changes in strain).

The appropriate linearized equations of motion for the independent variables \( s, u_i, \rho, \) and \( v_i \) are

\[
\partial_t s + \partial_t f_i = 0, \tag{6}
\]

\[
\partial_t u_i = U_i, \tag{7}
\]

\[
\partial_t \rho + \partial_t g_i = 0, \tag{8}
\]

\[
\partial_t g_i + \partial_t \Pi_{ik} = 0, \tag{9}
\]

where the fluxes \( f_i \) (of entropy), \( \Pi_{ik} \) (of momentum), \( g_i \) (of mass), and the “source” \( U_i \) (terminology introduced here) are given by

\[
f_i = s v_i - \frac{\kappa_{ij}}{T} \partial_j T - \frac{\alpha_{ij}}{T} \partial_j \lambda_{ij}, \tag{10}
\]

\[
U_i = v_i + \frac{\alpha_{ij}}{T} \partial_j T + \beta_{ij} \partial_j \lambda_{ij}, \tag{11}
\]

\[
\Pi_{ik} = \left( P \delta_{ik} - \lambda_{ik} \right) - \eta_{iklm} \partial_l v_m, \tag{12}
\]

\[
g_i = \rho v_i. \tag{13}
\]

AL use \( \sigma_{ik} \approx -\Pi_{ik} \) The term in (11) proportional to \( \beta_{ij} \) allows the lattice velocity \( \dot{u}_i \) to differ from the velocity \( v_i \) associated with mass flow. It leads, as we show, to a lattice diffusion mode for which \( \dot{u}_i \neq v_i \) and neither is zero.

Both the \( \alpha_{ij} \) and \( \beta_{ij} \) terms can be rewritten as flux terms. Linearizing about equilibrium, with primes denoting deviations from equilibrium, yields

\[
\partial_t u_i + \partial_j S_{ij} = v'_i, \tag{14}
\]
where
\[ S_{ij} = -\frac{\alpha_{ij} T'}{T} - \beta_{ij} \lambda^j, \] (15)

In (15), \( v_i \) can be thought of as a “lattice source”, and \( S_{ij} \) as a “lattice flux.” The \( \beta_{ij} \) term gives, in principle, anisotropic vacancy diffusion.

Recall that a diffusion constant \( D \) is proportional to a characteristic velocity times a characteristic mean-free path, so it has units of \( m^2/\text{sec} \). In terms of a \( D \), the dissipative coefficients have the following units: \( \kappa_{ij} \) has units of \( s \) times \( D \); \( \alpha_{ij} \) has units of \( D \); \( \beta_{ij} \) has units of inverse pressure times \( D \); and \( \eta_{iklm} \) has units of \( \rho \) times \( D \).

### III. CRYSTAL UNDER PRESSURE

#### A. Internal Pressure and Elasticity

The momentum conservation equation (9) implicitly contains the term \( \lambda_{ik} = -P \delta_{ik} \), which determines the force on the surface of the solid. An internal pressure \( P \) does not appear in the thermodynamics of Ref. 31, which does not consider either a lattice under applied pressure or the presence of defects. However, the extensive energy \( E = e V \), which depends on the extensive variables \((S, V, N, W_{ik} \equiv W_{ik}, V, \bar{g})\), has second derivatives that satisfy the Maxwell relation
\[ -\frac{\partial P}{\partial W_{ik}} = \frac{\partial \lambda_{ik}}{\partial V}, \] (16)

where the appropriate variables are held constant. For solid \( ^{4}\text{He} \) under an applied pressure \( P_a \), this makes \( P \) non-zero.

In principle we may let \( E \) depend on the number of vacancies \( N_V \), with associated “chemical potential” \( \phi_V = \partial E/\partial N_V \) (with units of energy, rather than velocity squared). Then the additional Maxwell relation
\[ -\frac{\partial P}{\partial N_V} = \frac{\partial \phi_V}{\partial V}, \] (17)

follows, with the appropriate variables held constant. If the vacancies are not in equilibrium (i.e., \( \phi_V \neq 0 \)), this also makes \( P \) non-zero. The general results of the present work (e.g., a nonzero lattice diffusion constant) can thus be made applicable to a solid not under \( P_a \) but having a large number of local thermal equilibrium. Terms found here to depend on \( P_a \) may in that case depend on the difference between actual concentration of vacancies and the equilibrium concentration of vacancies. However, we expect a \( P_a \) of \( \sim 25 \text{ atm} \) to dominate the effect of vacancies, and thus neglect their effect on \( P \). Although Refs. 1, 26 and 28 introduce the internal pressure \( P \), they do not calculate \( P \) or its thermodynamic derivatives. Ref. 1 and the present work neglect the possibility of interstitial atoms. For a reference that considers interstitials, see Ref. 33.

As employed by Ref. 1, this pressure term, in contrast to \( \lambda_{ik} \) alone (Ref. 31 does not include \( P \)), permits one to continuously approach the superfluid limit, when the lattice disappears. In the present case, it permits one to continuously approach the ordinary liquid limit.

The consequences of a nonzero \( P \) include, but are not limited to, a mode where vacancies are permitted to diffuse. Thermodynamic derivatives of \( P \) are essential for defect diffusion, and also affect the elastic modes. Moreover, they are needed to obtain the pure liquid limit for longitudinal sound on letting the crystallinity go to zero. We first use a Maxwell relation to find an explicit expression for \( P \) as a function of strain.

#### B. Internal Pressure \( P \)

Since holding \((V, N)\) constant is equivalent to holding \((V, \rho)\) constant, and similarly for \((S, N)\) and \((\sigma = s/\rho, N)\), we use these sets interchangeably. We rewrite (10) as
\[ -\frac{\partial P}{\partial W_{ik}} = \frac{\partial \lambda_{ik}}{\partial V}. \] (18)

For constant \( W_{ik} \) we have
\[ 0 = dW_{ik} = w_{ik} dV + V dw_{ik}, \] (19)

so that
\[ dw_{ik} = \frac{w_{ik}}{V}. \] (20)

Then
\[ \frac{\partial \lambda_{ik}}{\partial V} \bigg|_{W_{ik}, S, N} = \frac{\partial \lambda_{ij}}{\partial V} \bigg|_{w_{ik}, \sigma, N} - \frac{w_{jl}}{V} \frac{\partial \lambda_{ik}}{\partial w_{jl}} \bigg|_{V, \sigma, \rho}, \] (21)

and (18) gives
\[ \frac{\partial P}{\partial w_{ik}} = -V \frac{\partial \lambda_{ik}}{\partial V} \bigg|_{w_{ik, S, N}} = -V \frac{\partial \lambda_{ij}}{\partial V} \bigg|_{w_{ik, \sigma, N}} + \frac{w_{jl}}{V} \frac{\partial \lambda_{ik}}{\partial w_{jl}} \bigg|_{V, \sigma, \rho}. \] (22)

We employ Ref. 31 for the elasticity tensor \( \lambda_{ik} \) in an isotropic solid. Using superscript (0) to denote the equilibrium value of \( \lambda_{ik} \) and the strain \( w_{ik} \), we have
\[ \lambda_{ik}^{(0)} = \left(K - \frac{2}{3} \mu \right) \delta_{ik} w_{ij}^{(0)} + \mu V \left( w_{ik}^{(0)} + w_{ki}^{(0)} \right), \] (23)

where \( K \) and \( \mu \) are the bulk and shear moduli, and both \( \lambda_{ik}^{(0)} \) and \( w_{ik}^{(0)} \) are to be determined under a given applied pressure \( P_a \). Eq. (22) then gives
\[ \frac{\partial P}{\partial w_{ik}} \bigg|_{V, \sigma, \rho} = \left(K^* - \frac{2}{3} \mu^* \right) \delta_{ik} w_{ij}^{(0)} + \mu^* \left( w_{ik}^{(0)} + w_{ki}^{(0)} \right), \] (24)
where
\[ K^* = K - V \left. \frac{\partial K}{\partial \nu} \right|_{w_{ik}, \sigma, N}, \quad \mu^*_V = \mu_V - V \left. \frac{\partial \mu_V}{\partial \nu} \right|_{w_{ik}, \sigma, N}. \]

Under uniform \( P_a \) we expect an isotropic response, so
\[ w_{ik}^{(0)} = \frac{\delta_{ik}}{3} w_{ll}^{(0)}. \]  

Then (24) becomes
\[ \frac{\partial P}{\partial w_{ik}} \bigg|_{V, \sigma, \rho} = K^* \delta_{ik} w_{ll}^{(0)}. \]  

Integration of (27) with respect to \( w_{ik} \) gives the part of the internal pressure dependent on the strain to be
\[ P = \frac{1}{2} K^* \left( w_{ll}^{(0)} \right)^2, \]
where we take the integration constant to be zero. For \( w_{11}^{(0)} = w_{22}^{(0)} = w_{33}^{(0)} \), we then have
\[ P = \frac{9}{2} K^* w_{11}^{(0)}^2. \]

This result applies to the case of a strongly crystalline material. In the opposite limit where the crystallinity disappears and the particles are weakly interacting, part of \( P \) would be given by the ideal gas law.

**C. Strain \( w_{ik} \)**

As discussed above, under an applied pressure the force on the surface of a solid is
\[ \lambda_{ik}^{(0)} = P \delta_{ik} = -P_a \delta_{ik}. \]

Taking the trace yields
\[ \frac{\lambda_{ll}^{(0)}}{3} - P = -P_a. \]

Substitution from (23) and (24) gives
\[ 3K w_{11}^{(0)} - \frac{9}{2} K^* w_{11}^{(0)}^2 = -P_a. \]

Since an applied pressure should cause a negative strain, only the solution for \( w_{11}^{(0)} < 0 \) is physical.

For solid \(^4\)He, we expect both \( w_{11}^{(0)} \) and \( P_a/K \) to be small. The solution of (32) to second order in \( P_a \) is
\[ w_{11}^{(0)} \approx -\frac{P_a}{3K} + \frac{P_a^2 K^*}{6K^3}. \]

The first term is what one would get on neglecting \( P \) in (31). To second order in \( P_a/K \), eq. (29) then gives
\[ P = K^* \frac{P_a^2}{2K^2}. \]

**IV. NORMAL MODES OF ANDREEV-LIFSHITZ NORMAL SOLID WITH DEFECTS**

As noted earlier, this system has eight variables: \( s, \rho, g_i \) and \( u_i \). Disturbances from equilibrium will be denoted by primes, so we use \( s', \rho', g'_i \approx \rho v'_i \), and \( u'_i \). There are correspondingly eight normal modes. For an infinite system we assume a disturbance of the form \( \exp[i(\vec{k} \cdot \vec{r} - \omega t)] \), where the real wavevector \( \vec{k} \) is considered to be known, but \( \omega \) is unknown. For the disturbance to decay in time, \( S(\omega) < 0 \). Six modes come in three degenerate pairs, with \( g'_i \) and \( u'_i \) strongly coupled, and correspond to ordinary elasticity. The other two modes are diffusive, with temperature diffusion nearly decoupled from lattice diffusion. To ensure this decoupling we set the (off-diagonal) temperature-lattice transport coefficient \( \alpha_{ij} \) to be 0, and set the distinct but similar-looking thermal expansion coefficient \( \beta_{ij} \) to be 0. We consider an isotropic solid, for which \( \kappa_{ij} = \kappa \delta_{ij} \) and \( \beta_{ij} = \beta \delta_{ij} \), this \( \beta \) not to be confused with the identical symbol sometimes used for the thermal expansion coefficient \( \beta \).

We also neglect the tensor viscosity \( \eta_{iklm} \), which to lowest order in \( k \) does not contribute to the modes. The fluctuation of the tensor \( \Pi_{ik} \) (12) has a term from the viscosity \( \sim \eta_{iklm} k_m v'_l \) and a term from the stress tensor \( \sim \lambda_{ik}^{(0)} = (\partial \lambda_{ik}/\partial w_{jl}) w_{jl}' \). Thus, \( \lambda_{ik}^{(0)} \sim \omega' k^2 \), and \( \lambda_{ik}^{(0)} \sim k^2 / \omega \). Thus, for both propagating modes (\( \omega \sim k \)) and diffusive modes (\( \omega \sim k^2 \)), the term in \( \Pi_{ik} \) due to viscosity is, at the least, of order \( k \) relative to the term \( \lambda_{ik}^{(0)} \), and is therefore neglected in the long wavelength limit.

**A. Thermal Diffusion**

For the normal solid it is convenient to work with \( \rho \) and \( \sigma = s/\rho \) as variables, because \( \sigma \) diffuses but does not flow, and therefore is nearly conserved. To see this note that, to lowest order in deviations from equilibrium, eqs. (4) and (8) yield
\[ \partial_t \sigma' = \frac{1}{\rho} \partial_t \left( \frac{\kappa}{T} \partial_T T \right) \approx \frac{\kappa}{T(\partial \sigma/\partial T)_{\rho}} \nabla^2 \sigma', \]
where we have used \( \alpha = 0 \). This equation describes entropy diffusion, with \( \sigma' \neq 0 \) and
\[ \omega = -i D_T k^2, \quad D_T = \frac{\kappa}{\rho T(\partial \sigma/\partial T)_{\rho}}. \]
For this mode $u'_i = v'_i = \rho' = 0$. If $\alpha$ is small but non-zero the frequency will not change to lowest order in $\alpha$, but from the equations for $\rho$, $\tilde{g}$, and $\bar{u}$ these quantities would develop amplitudes proportional to $\alpha'$ and $\alpha$, and thus have negligible amplitude as $\alpha \to 0$. We consider only the case where the effects of $\alpha$ can be neglected.

### B. Elastic Modes

We obtain the elastic modes by taking $\sigma' = 0$ and neglecting dissipative and nonlinear terms in (7)-(9). Thus, eq. (7) gives $\dot{u}'_i = v'_i$. In the remainder of this work, all thermodynamic derivatives are taken at constant $\sigma$, and derivatives with respect to $\rho$ are taken at constant $w_{ik}$ and vice-versa, unless otherwise specified. Further, when derivatives with respect to a specific component of $w_{ik}$ are taken, the other components of $w_{ik}$ are held fixed. Then by (13) and (19), eqs. (38) and (39) become\(^{39}\)

\[
0 = \rho \ddot{u}'_i + \frac{\partial P}{\partial \rho} \partial_{\rho} \rho' - \frac{\partial P}{\partial w_{ij}} \partial_{w_{ij}} w_{ij}' - \frac{\partial \lambda_{ik}}{\partial \rho} \partial_{\rho} \partial_{k} \rho' - \frac{\partial \lambda_{ik}}{\partial w_{ij}} \partial_{w_{ij}} \partial_{k} w_{ij}' ,
\]

\[(38)\]

\[
0 = \ddot{\rho}' + \rho \partial_{\rho} \dot{u}'_i .
\]

\[(39)\]

Clearly, $\sigma'$ does not couple to the other variables. On linearizing, eq. (39) gives $\rho' = -\rho \partial_{\rho} u'_i$, so with (2), eq. (39) becomes\(^{38}\)

\[
0 = \rho \ddot{u}'_i - \frac{\partial P}{\partial \rho} \partial_{\rho} w_{ik}' + \frac{\partial P}{\partial w_{ij}} \partial_{w_{ij}} j' \partial_{j} j' \partial_{w_{ij}} \partial_{j} j' - \frac{\partial \lambda_{ik}}{\partial \rho} \partial_{\rho} \partial_{k} \partial_{w_{ij}} \partial_{w_{ij}} \partial_{k} w_{ij}' .
\]

\[(40)\]

The second term gives the pure fluidlike (longitudinal) response, which occurs for $P \neq 0$ (e.g., an imperfect solid or a solid under $P_a$), and the fifth term gives the pure solidlike (longitudinal and transverse) response.

Appendix B shows that, for uniform static $P_a$, certain quantities are isotropic. This permits us to define

\[
\frac{\partial P}{\partial w_{ij}} = \frac{\partial P}{\partial w} \delta_{ij}, \quad \frac{\partial \lambda_{ij}}{\partial \rho} = \frac{\partial \lambda}{\partial \rho} \delta_{ij}, \quad \frac{\partial \lambda}{\partial w} \equiv K = \frac{4}{3} \mu V .
\]

\[(41)\]

Appendix B also shows that

\[
\frac{\partial \lambda_{ik}}{\partial w_{ij}} \bigg|_{\rho, \sigma} = \frac{\partial \lambda}{\partial w} \delta_{ik} \delta_{ij} + \mu V (\delta_{ij} \delta_{kl} + \delta_{kij} \delta_{il} - 2 \delta_{ik} \delta_{jl}) .
\]

\[(42)\]

Thus (40) gives

\[
0 \approx \rho \ddot{u}'_i - \frac{\partial P}{\partial \rho} \partial_{\rho} w_{ik}' + \frac{\partial P}{\partial w} \partial_{w} \partial_{k} w_{ik}' \\
+ \frac{\partial \lambda}{\partial \rho} \partial_{\rho} \partial_{k} w_{ik}' - \left( \frac{\partial \lambda}{\partial w} - \mu V \right) \partial_{w} \partial_{r} u'_i - \mu V \nabla^2 u'_i .
\]

\[(43)\]

On letting $\partial_i \to ik$ and $\partial_i \to -i\omega$, eq. (13) becomes

\[
0 \approx (-\rho \omega^2 + \mu V) k^2 u'_i \\
+ \left[ \frac{\partial P}{\partial \rho} - \frac{\partial P}{\partial w} \omega + \frac{\partial \lambda}{\partial \rho} - \mu V \right] k_i (\vec{k} \cdot \vec{u}) .
\]

\[(44)\]

**Longitudinal Mode:** If $\vec{k} \cdot \vec{u} = 0$, then (44) shows that $u_i$ is along $k_i$, so the mode is longitudinal. Moreover, eq. (44) gives the normal mode frequencies

\[
\omega^2 = \left[ \frac{\partial P}{\partial \rho} - \frac{1}{\rho} \frac{\partial P}{\partial w} \omega + \frac{\partial \lambda}{\partial \rho} + \frac{1}{\rho} \frac{\partial P}{\partial w} \right] k^2 \\
= \left[ c_{L}^2 + c_{S}^2 \right] k^2 \equiv c_{L}^2 k^2 .
\]

\[(45)\]

where

\[
c_{L}^2 = \frac{\partial P}{\partial \rho} - \frac{\partial \lambda}{\partial \rho} , \quad c_{S}^2 = \frac{1}{\rho} \frac{\partial P}{\partial w} - \frac{1}{\rho} \frac{\partial P}{\partial w} .
\]

\[(46)\]

The liquid-like velocity $c_{L}$ contains thermodynamic derivatives with respect to the density $\rho$, and the solid-like velocity $c_{S}$ contains thermodynamic derivatives with respect to the strain $w_{ik}$. Eq. (45) gives a velocity for longitudinal sound that is similar to that found in Ref. 28.

Appendix B finds the four derivatives in (46) in terms of $P_a$, which to second order in $P_a/K$ give

\[
c_{L}^2 = \frac{P_a}{\rho} \left( \frac{K^*}{K} - 1 \right) + \frac{P_a^2 K^*}{2 \rho K^2} \left( 1 - \frac{K^*}{K} + \rho \frac{\partial K^*}{\partial \rho} \right) ,
\]

\[(47)\]

\[
c_{S}^2 = \frac{K + \frac{4}{3} \mu V}{\rho} + \frac{P_a K^*}{\rho} - \frac{P_a^2 K^*}{2 \rho K^2} .
\]

\[(48)\]

where $K^*$ is defined in (25). For $P_a = 0$ we have $c_{L}^2 = c_{S}^2 = [K + (4/3) \mu V]/\rho$, which agrees with Ref. 31 for an ordinary solid.

**Transverse Mode:** If $\vec{k} \cdot \vec{u} = 0$, so that the mode is transverse, then (44) gives the normal mode frequencies

\[
\omega^2 = \frac{\mu V}{\rho} k^2 .
\]

\[(49)\]

From (39), for the transverse mode $\rho' = 0$. Eq. (49) agrees with Ref. 31 for an ordinary solid.

For both longitudinal and transverse mode frequencies, eq. (49) is satisfied by $\sigma' = 0$.

### C. Lattice Diffusion

The lattice diffusion mode is the most subtle of the modes. For this mode, as for the elastic modes, we consider that $\sigma$ is constant, but we do not take $v'_i = u'_i$. Rather, we assume that $\omega = -iD_{L} k^2$, where the lattice mode diffusion constant $D_{L}$ is to be determined, and we keep the dissipative terms in the equations of motion for $v'_i$, $u'_i$, and $\rho'$.
With $\beta_{ij} = \beta \delta_{ij}$ (i.e., an isotropic solid), $\alpha_{ij} = 0$, and setting $\sigma' = 0$, eqs. (5) and (7) give

$$-i\omega \rho' = -\rho(ik_i)v'_i, \quad (50)$$
$$-i\omega u'_i = v'_i + \beta(ik_i)\lambda'_i. \quad (51)$$

If we assume that the mode is longitudinal, with $v'_i \sim k_i$ (the consistency of this assumption to be determined below), then the first of these equations implies that $v'_i \sim k_i \rho'$. Therefore in (50) the term $\partial / \partial P$ is of order $k^2$ relative to the $k \rho'$ dependence of $\partial \lambda / \partial \rho$, and is neglected in the long wavelength limit. As a consequence, $\partial / \partial \rho$ is replaced by $\partial / \partial P$. Thus, neglecting the $\partial / \partial \rho$ term and neglecting the viscosity $\eta_{iklm}$ (as discussed above), eq. (51) gives

$$ik_i \rho \frac{\partial P}{\partial \rho} - ik_i \rho \frac{\partial \lambda_{ik}}{\partial \rho} = -k_k k_j \partial / \partial w_{ji} \lambda_{ki} + \frac{\partial P}{\partial w_{ji}} k_k k_j u'_i. \quad (52)$$

Substitution from (51) and (52) gives

$$ik_i \rho \frac{\partial P}{\partial \rho} - ik_i \rho \frac{\partial \lambda_{ik}}{\partial \rho} = -k_k k_j \partial / \partial w_{ji} \lambda_{ki} + \frac{\partial P}{\partial w_{ji}} k_k k_j u'_i. \quad (53)$$

All but one term in (53) is along $k_i$, and the remaining term is along $u'_i$. Therefore we deduce that $u'_i$ is along $k_i$, and thus $k_i k_i u'_i = k^2 u'_i$. Then (53) becomes

$$ik_i \rho \frac{\partial P}{\partial \rho} - ik_i \rho \frac{\partial \lambda_{ik}}{\partial \rho} = -k_k k_j \frac{\partial \lambda_{ki}}{\partial w_{ji}} u'_i + \frac{\partial P}{\partial w_{ji}} k_k k_j u'_i. \quad (54)$$

Further, eq. (51) gives, on taking $\lambda'_{ik} = (\partial / \partial w_{ji}) w_{ji} + (\partial / \partial \rho) \rho'$, and taking $u'_i$ along

$$-i\omega + \beta \frac{\partial \lambda}{\partial w} k^2 u'_i = v'_i + ik_i \beta \frac{\partial \lambda}{\partial \rho} \rho'. \quad (55)$$

Since $u'_i$ is along $k_i$, eq. (55) implies $v'_i$ also along $k_i$. Hence the mode is longitudinal.

We now use (51) and the sound velocities of (10) to eliminate $\rho'$ from (54) and (55). Then (54) multiplied by $\omega$ gives

$$ipk_i \rho' \frac{\partial P}{\partial \rho} = -\omega k_i u'_i + \rho c^2_{1S} \frac{\partial P}{\partial \rho}. \quad (56)$$

and (55) multiplied by $\omega$ gives

$$-ipk_i \rho' \frac{\partial \lambda}{\partial w} k^2 = \omega + i\beta \rho \frac{\partial \lambda}{\partial \rho} k^2 \quad (57)$$

Since $u'_i$ and $v'_i$ are along $k_i$, eq. (56) implies that for the diffusive mode

$$v'_i = i\omega u'_i (c^2_{1S} / c^2_{1L}) = -u'_i (c^2_{1S} / c^2_{1L}), \quad (58)$$

which is independent of $\omega$. We interpret this as the lattice velocity $u'_i$ being out of phase relative to the matter velocity $v'_i$ so that the fluid and lattice stresses cancel. Combining (56) and (57) then yields

$$\omega (c^2_{1S} + c^2_{1L}) = -ik^2 \beta \rho \left[ \frac{3 \partial \lambda}{c^2_{1S} \partial \rho} + \frac{2 \partial \lambda}{c^2_{1L} \rho} \right] \frac{\partial P}{\partial \rho}.$$
that the experimental results of Ref. 22 are due to a lattice diffusion mode leads to the conclusion that \( \tau \) varies as \( \exp[\Delta/k_B T] \). Indeed, such a dependence is observed, with \( \Delta \sim 30 \text{ mK} \). It would be useful to test for the predicted \( \phi^2 \)-dependence. For instance, the present theory predicts that changing the plate separation in the pancake cell of Ref. 22 from 100 \( \mu \text{m} \) to 200 \( \mu \text{m} \) should yield a relaxation time approximately four times longer.

This mode provides a means for vacancy flow to equilibrate vacancy concentrations. It is consistent with the observation of Ref. 22 that pressure decreases during an anneal, and when the system relaxes at constant temperature. We interpret this to mean that vacancies diffuse to or from the surface.

We now turn to how a normal solid will respond to the two devices usually employed to generate a disturbance: a heater and a transducer. Since there are three longitudinal modes (thermal diffusion, lattice diffusion, and elastic waves), it would appear that there is need for an additional independent generator. Perhaps surface properties introduce a new boundary condition that amounts to having an independent generator. For example, the material against the solid \(^4\text{He}\) may cause the \(^3\text{He}\) surface to prefer vacancies, as opposed to atoms. Thus the surface treatment may affect the behavior of both heaters and transducers. This argument applies to any two ordinary solids, and there may be some for which this can be readily tested. Hence two macroscopically identical heaters or transducers made of different materials, or of the same material but with different surface treatment, would not show identical behavior. Since \( v_i - \dot{u}_i \approx 0 \) for the temperature mode and the elastic modes, one way to characterize the response of a surface is in terms of \( v_i - \dot{u}_i \). Thus \( (v_i - \dot{u}_i)/T \) for a longitudinally moving transducer and \( (v_i - \dot{u}_i)/T' \) for a heater would characterize differences in the response to different surface conditions, and the extent to which they can generate the lattice diffusion mode.

\section{SUMMARY AND CONCLUSIONS}

We have applied the Andreev-Lifshitz theory of supersolid dynamics to an ordinary solid with lattice defects specifically, with vacancies in mind. At the thermodynamic level, this theory includes an internal pressure \( P \), distinct from the applied pressure \( P_a \) and the stress tensor \( \lambda_{ik} \). For the Andreev-Lifshitz theory this is necessary to permit a continuous variation from a supersolid to a superfluid. Under uniform static \( P_a \), we have \( \lambda_{ik} = (P - P_a)\delta_{ik} \). For \( P_a \neq 0 \), Maxwell relations imply that \( P \sim P_a \). These results are not conventional; Ref. 31 does not include \( V \) as a distinct extensive thermodynamic variable, nor its thermodynamically conjugate variable \( P \). In the present work many derivatives involving \( V \) are at fixed strain \( w_{ik} \), which is also unconventional, since normally one assumes that \( \delta w_{ii} = -\delta \rho/\rho \). Nevertheless, the variables of Andreev and Lifshitz must be taken if vacancies are to be permitted.

For an isotropic model, the normal modes were obtained. There are, as expected, two sets of propagating transverse modes, with velocities as expected. There also are, as expected, a set of propagating longitudinal modes, but with velocities containing both solid-like and liquid-like contributions, and which depend upon \( P_a \). In addition there are two diffusive longitudinal modes: a well-known mode that dominantly involves temperature, and another mode involving lattice defects (i.e., vacancies). Our analysis of the physical nature of this mode shows that it is surprisingly complex. It involves the mass density \( \rho \), the lattice velocity \( \dot{u}_i \), and the mass-flow velocity \( v_i \), with the fluid-like pressure \( P \) associated with \( \rho \) essentially canceling the solid-like stress \( \lambda \) associated with \( u_i \).

In a separate work 42 we discuss the normal modes of the full Andreev and Lifshitz theory for a supersolid, which has nine variables. As Ref. 1 established at \( T = 0 \), there are four pairs of propagating modes. Three pairs are essentially the elastic modes we have studied here, with a weak coupling to the superfluid. The fourth pair is basically a fourth sound mode, where the normal fluid is entrained by the lattice. These propagating modes, in the presence of a finite \( P_a \), and their generation by transducers and heaters, have been considered in Ref. 43. We also find a rather complex additional mode, not considered in Ref. 1, which is diffusive. Although the additional supersolid diffusive mode is similar to the normal solid diffusive mode found in the present work (e.g., zero net stress, and distinct mass and lattice motion), its mode structure differs significantly. The supersolid diffusive mode is characterized by three velocities: \( v_i' \) and \( u_i' \), and \( u_i' \), associated respectively with the normal mass, superfluid mass, and the lattice. For supersolid \(^4\text{He}\) with \( P_a \ll K \), we find that \( v_i' = v_i' \) and \( u_i' = u_i' \). We also find that \( g' = \rho_i v_i' + \lambda_i u_i' = 0 \). If \(^4\text{He}\) is a genuine supersolid, then this mode provides an alternate explanation for the exponential time-dependence of the pressure decay observed by Ref. 22.

We close with the following comment. Ref. 1 predicted that supersolidity will occur because of quantum diffusion, a situation that occurs at such low temperatures that the relevant bulk diffusion processes are temperature-independent. Ref. 22 observe temperature-dependent relaxation; therefore their system is not at a low enough temperature to be in the quantum diffusive regime. Note that quantum spin tunneling is an established phenomenon, wherein the magnetic relaxation rate saturates at low enough temperatures 44–46.

\section{ACKNOWLEDGEMENTS}

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Ref. 22 finds equations of motion identical to those of Ref. 21, but also includes nonlinear terms.

M. Liu, Phys. Rev. B 18, 1165 (1978). This work notes that Ref. 22 employs the non-Galilean \( \dot{J}_s = \rho_s \ddot{v}_s \) in place of the Galilean \( J_s = \rho_s (\dot{v}_s - \ddot{v}_s) \). This does not affect the equations of motion until the normal modes are calculated.


J. Bardeen and C. Herring, Imperfections in Nearly Perfect Crystals, John Wiley and Sons, Inc., New York, N. Y., 1952, p 261. This work treats a metal as a bicomponent system, with atoms and vacancies as the two components. It notes that mass motion relative to the lattice is not an assured result of including vacancies, as long as the vacancies are in local thermal equilibrium.


The present work uses the notation of Ref. 24, which follows Ref. 2 for the dissipative coefficients. On the other hand, Ref. 27 seems to use the notation of Ref. 24 for the dissipative coefficients, but follows Ref. 2 in using \( \lambda \) for the momentum density. The present work, Ref. 26, Ref. 24, and Ref. 26 use \( \dot{\mu} \) for the momentum density.

If \( N_L \) is the number of lattice sites and \( N_{os} \) is the number of on-site atoms, then \( N_L = N_{os} + N_{V} \). In addition, if there are \( N_I \) interstitial atoms, then \( N = N_{os} + N_I \). With the energy differential taking the form

\[
\frac{dE}{d\lambda} = \sum_i \phi_i dN_{Li} + \phi_{os} dN_{os} + \phi_{V} dN_{V}.
\]

we then have

\[
\frac{dE}{d\lambda} = \sum_i \phi_i dN_{Li} + \phi_{os} dN_{os} + \phi_{V} dN_{V} = \phi_i dN_{Li} + \phi_{os} dN_{os} + \phi_{V} dN_{V}.
\]

If the on-site and interstitials are in equilibrium, then both \( \phi_{os} = 0 \) and \( \phi_{V} = 0 \). We do not consider interstitials and only consider the case where \( P_s \) dominates the effect on \( P \).


In typical treatments of elasticity in solids it is implicit that \( P = 0 \), even for \( P_s \neq 0 \). To produce \( P = 0 \), even for \( w^{(0)} \neq 0 \), the present application of AL theory requires \( \lambda = 0 \). For that to hold, by eq. 22, \( \lambda \) must vary linearly with \( V \) at fixed strain \( \omega \); this appears to be unlikely.


See section 50 of Ref. 26.

In the linear approximation the corresponding two equations of Ref. 21 (see its eq.(19)), on taking \( P_s \to 0 \), agree with the above two equations.

To obtain agreement with Ref. 27, we drop higher-order terms in the velocity, take \( T = 0 \) (as in the present work), and neglect the static strain in eq. 3, which gives

\[
\frac{dP}{d\mu} \approx p_{ij} = \frac{\rho}{2} \left( \frac{\partial \lambda}{\partial \mu} \right)_{\rho = 0}.
\]

Then \( C^{ij}_{2} \) agrees with \( C^{ij}_{2} \) found in Ref. 22, and \( D_{ij} \) of 26 agrees with \( D_{ij} \) found in Ref. 27. However, for finite \( P_s \) it is inconsistent to neglect the static strain (i.e., \( \rho(\partial\lambda/\partial\mu) \sim \rho(\partial\lambda/\partial\mu) \approx (P_s/K)^2 \approx (\Delta\lambda_s/\rho_{ij}) \)).

To obtain agreement with Ref. 26, we take \( \partial\lambda/\partial\mu \) to dominate the denominator of 26, and make the identifications, valid for zero static strain (such as \( P_s = 0 \) and vacancies in equilibrium), that

\[
\rho(\partial\lambda/\partial\mu) \rightarrow \gamma R \text{ and } \rho(\partial\lambda/\partial\mu) \rightarrow \chi R/4,
\]


M. Sears and W. M. Saslow, “Generation Efficiencies for Longitudinal Propagating Modes in a Supersolid”, accepted by Physical Review B.


The term \( -u_i \partial_j v_{ij} \), proportional to the lattice position \( u_i \), would cause \( u_i \) to depend upon the choice of origin; this is not translationally invariant.
Appendix A: Andreev-Lifshitz Supersolid

1. Thermodynamics

Consider a general frame of reference, with non-zero superfluid velocity \( \vec{v}_n \) and normal fluid velocity \( \vec{v}_s \). Let \( \vec{u} \) be the local displacement of the crystal sites relative to their equilibrium, and take the strain to be given by \( w_{ik} = \partial_i u_k \). Then by thermodynamics the differential of the energy density \( \epsilon \) is given by

\[
d e = T d s + \lambda_{ik} d w_{ik} + \mu d \rho + \bar{\gamma}_{s} \cdot d \vec{v}_s + \bar{\gamma}_{n} \cdot d \vec{g}.
\]

(A1)

Here \( \lambda_{ik} \) is an elastic tensor density (with the same units as pressure \( P \)), \( \mu \) is the chemical potential (with units of velocity squared), \( \bar{\gamma}_{s} = \vec{g} - \rho \vec{v}_n \) (a requirement of Galilean relativity), \( \rho = \rho_n + \rho_s \) (the sum of the normal and superfluid densities), and \( \bar{\gamma} = \rho_n \vec{v}_n + \rho_s \vec{v}_s \). By thermodynamic extensivity we also have

\[
\epsilon = -P + T s + \lambda_{ik} w_{ik} + \mu \rho + \bar{\gamma}_{s} \cdot \vec{v}_s + \bar{\gamma}_{n} \cdot \vec{g} \tag{A2}
\]

and the Gibbs-Duhem relation

\[
0 = -dP + s dT + w_{ik} d \lambda_{ik} + \rho d \mu + \bar{\gamma}_{s} \cdot d \vec{v}_s + \bar{\gamma}_{n} \cdot d \vec{g} . \tag{A3}
\]

The system will be in equilibrium when the thermodynamic forces \( \partial_i T, \partial_i \lambda_{ik}, \partial_i \mu, \partial_i v_{nj}, \) and \( \partial_i j_{ai} \) are all zero.

2. Dynamics

The thermodynamic variables \( \epsilon, s, u_i, \rho, \vec{v}_s, \) and \( \vec{g} \) are taken to satisfy equations of motion that are first order in time and that satisfy appropriate properties under space rotation and inversion, and under time-reversal. Thus \( \epsilon, s, \rho, \) and \( \vec{g} \) satisfy conservation laws (a flux but no source), the phase gradient \( \vec{v}_s \) is proportional to a gradient (a type of flux, with no source), and the displacement \( u_i \) has a source but no flux. Thus

\[
\partial_t \epsilon + \partial_i Q_i = 0, \tag{A4}
\]

\[
\partial_t s + \partial_i f_i = \frac{R}{T}, \quad (R \geq 0), \tag{A5}
\]

\[
\partial_t u_i = U_i, \tag{A6}
\]

\[
\partial_t \vec{v}_s + \vec{V} \theta = 0, \tag{A7}
\]

\[
\partial_t \rho + \partial_i g_i = 0, \tag{A8}
\]

\[
\partial_t g_i + \partial_k \Pi_{ik} = 0. \tag{A9}
\]

(The source \( U_i \) was implicit in previous theories\(^{123}\)). The unknown fluxes \( Q_i, f_i, \phi, \) and \( \Pi_{ik} \), and the unknown sources \( R \) and \( U_i \), are determined by subjecting them to the condition that, when applied to the thermodynamic equation (A1), the density \( R \) of the rate of dissipated energy be non-negative. Note that \( g_i \) is already known, and \( Q_i \) and \( R \) will not be needed. For \( f_i, U_i, \theta, \) and \( \Pi_{ik} \) we have, when terms non-linear in velocities and strains are neglected,

\[
f_i = s v_{ni} - \frac{\kappa_{ij}}{T} \partial_j T - \frac{\alpha_{ij}}{T} \partial_i \lambda_{ik}, \tag{A10}
\]

\[
U_i = v_{ni} + \frac{\alpha_{ij}}{T} \partial_j T + \beta_{ij} \partial_i \lambda_{ik}, \tag{A11}
\]

\[
\theta = \mu - \zeta_{ik} \partial_k v_{ni} - \chi_{ik} j_{sk}, \tag{A12}
\]

\[
\Pi_{ik} = (P \delta_{ik} - \lambda_{ik}) - \eta_{iklm} \partial_m v_{nl} - \zeta_{ik} j_{sk}. \tag{A13}
\]

In each of these equations, the last two terms are dissipative and the preceding terms are reactive.

Refs. \(^1\) and \(^{23}\) obtain a nonlinear term in \( U_i \), which may be obtained by letting \( v_i \rightarrow v_i - v_j \partial_j u_i \). On the other hand, Ref. \(^{27}\) obtains two nonlinear terms, which may be obtained by letting \( v_i \rightarrow v_i - v_j \partial_j u_i - u_i \partial_j v_j \).

Appendix B: Relevant Thermodynamic Derivatives

In what follows, the quantities \( \frac{\partial P}{\partial w} \rho, \sigma \), \( \frac{\partial P}{\partial w} \rho, \sigma_{ik} \), \( \frac{\partial \lambda_{ik}}{\partial \rho} \rho, \sigma \), and \( \frac{\partial \lambda_{ik}}{\partial \rho} \rho, \sigma_{ik} \) are obtained in terms of \( P_a \) and the elastic constants.

(1) With \( w^{(0)}_{11} = w^{(0)}_{22} = w^{(0)}_{33} \), eq. \(^{27}\) gives

\[
\frac{\partial P}{\partial w_{ik}} \bigg|_{\rho,\sigma} = 3K^* \delta_{ik} w^{(0)}_{11} \equiv \frac{\partial P}{\partial w} \delta_{ik}. \tag{B1}
\]

where \( \frac{\partial P}{\partial w} \) is defined for later convenience. Substitution for \( w^{(0)}_{11} \) from \(^{23}\) gives, to second order in \( P_a/K \),

\[
\frac{\partial P}{\partial w} \approx - P_a K + \frac{P_a^2 K^*}{2K^3}. \tag{B2}
\]

(2) From \(^{29}\) we have

\[
\frac{\partial P}{\partial \rho} \bigg|_{\sigma, w_{ik}} = \frac{9}{2} w^{(0)}_{11} \frac{\partial K^*}{\partial \rho} \bigg|_{\sigma, w_{ik}}. \tag{B3}
\]

By \(^{26}\),

\[
\frac{\partial K^*}{\partial \rho} \bigg|_{\sigma, w_{ik}, N} = \frac{\partial}{\partial \rho} \left( \frac{K - V}{\partial \rho} \frac{\partial K^*}{\partial \rho} \bigg|_{\sigma, w_{ik}, N} \right) \bigg|_{\sigma, w_{ik}} \tag{B4}
\]

Thus \(^{30}\) can be written as

\[
\frac{\partial P}{\partial \rho} \bigg|_{\sigma, w_{ik}} = \frac{9V^2}{2} w^{(0)}_{11} \frac{\partial^2 K}{\partial \rho^2} \bigg|_{\sigma, w_{ik}, N}. \tag{B5}
\]

To second order in \( P_a/K \), eqs. \(^{30}\), \(^{31}\) and \(^{31}\) give

\[
\frac{\partial P}{\partial \rho} \bigg|_{\sigma, w_{ik}} \approx \frac{1}{2} \frac{P_a^2}{K^2} \frac{\partial K^*}{\partial \rho} \bigg|_{\sigma, w_{ik}} = \frac{V^2 P_a^2}{2K^2} \frac{\partial^2 K}{\partial \rho^2} \bigg|_{\sigma, w_{ik}, N}. \tag{B6}
\]
From (23) we have
\[
\frac{\partial \lambda_{ik}}{\partial w_{jl}}|_{\rho, \sigma} = \left(K - \frac{2}{3} \mu V\right) \delta_{ik} \delta_{jl} + \mu V (\delta_{ij} \delta_{kl} + \delta_{kj} \delta_{il}).
\]
(B7)

We now define
\[
\frac{\partial \lambda}{\partial w} \equiv K + \frac{4}{3} \mu V,
\]
so that
\[
\frac{\partial \lambda_{ik}}{\partial w_{jl}}|_{\rho, \sigma} = \frac{\partial \lambda}{\partial w} \delta_{ik} \delta_{jl} + \mu V (\delta_{ij} \delta_{kl} + \delta_{kj} \delta_{il} - 2 \delta_{ik} \delta_{jl}).
\]
(B9)

From (23) we also have
\[
\left. \frac{\partial \lambda_{ik}}{\partial \rho} \right|_{w_{ik}, \sigma} = \left. \left( \frac{\partial K}{\partial \rho} \right|_{w_{ik}, \sigma} - \frac{2}{3} \frac{\partial \mu V}{\partial \rho} \right) \delta_{ik} w_{ll}^{(0)} + \frac{\partial \mu V}{\partial \rho} \left( w_{ik}^{(0)} + w_{ki}^{(0)} \right).
\]
(B10)

With
\[
\frac{\partial K}{\partial \rho} \left|_{w_{ik}, \sigma} = - \frac{V}{\rho} \frac{\partial K}{\partial N} \right|_{w_{ik}, \sigma, N} = \frac{K^* - K}{\rho},
\]
(B11)

and with a similar relation for \(\mu V\), eq. (B10) gives
\[
\left. \frac{\partial \lambda_{ik}}{\partial \rho} \right|_{w_{ik}, \sigma} = \left( \frac{K^* - K}{\rho} - \frac{2}{3} \frac{\mu^* - \mu V}{\rho} \right) \delta_{ik} w_{ll}^{(0)} + \frac{\mu^* - \mu V}{\rho} \left( w_{ik}^{(0)} + w_{ki}^{(0)} \right).
\]
(B12)

With (26) and \(w_{11}^{(0)} = w_{22}^{(0)} = w_{33}^{(0)}\),
\[
\left. \frac{\partial \lambda_{ik}}{\partial \rho} \right|_{w_{ik}, \sigma} = \left( \frac{K^* - K}{\rho} \right) \delta_{ik} w_{ll}^{(0)} = 3 \left( \frac{K^* - K}{\rho} \right) \delta_{ik} w_{11}^{(0)}
\]
(B13)

where \(\partial \lambda/\partial \rho\) is defined for later convenience. To second order in \(P_a/K\), eq. (33) gives
\[
\frac{\partial \lambda}{\partial \rho} \approx \left( 1 - \frac{K^*}{K} \right) \left[ \frac{P_a}{\rho} - \frac{P_a^2 K^*}{2 \rho K^2} \right].
\]
(B14)