Ginzburg-Landau theory for the solid-liquid interface of bcc elements.
II. Application to the classical one-component plasma, the Wigner crystal, and ⁴He

X. C. Zeng and D. Stroud

Department of Physics, Ohio State University, Columbus, Ohio 43210

(Received 8 December 1988)

We extend our previously developed Ginzburg-Landau theory for calculating the crystal-melt interfacial tension of bcc elements to treat the classical one-component plasma (OCP), the charged fermion system, and the Bose crystal. For the OCP, a direct application of the theory of Shih *et al.* [Phys. Rev. A **35**, 2611 (1987)] yields for the surface tension $\tau = 1.12 \times 10^{-3} (Z^2 e^2 / a^3)$, where Ze is the ionic charge and a is the radius of the ionic sphere. For the fermion system, the absence of reliable correlation functions near the coexistence line makes it difficult to estimate the surface tension. We treat the Bose crystal-melt interface by a quantum extension of the classical density-functional theory, using the Feynman formalism to estimate the relevant correlation functions. The theory is applied to the metastable ⁴He solid-superfluid interface at $T = 0$, with a resulting surface tension $\tau \approx 0.085$ erg/cm², in reasonable agreement with the value extrapolated from the measured surface tension of the bcc solid in the range 1.46–1.76 K. These results suggest that the density-functional approach is a satisfactory mean-field theory for estimating the equilibrium properties of liquid-solid interfaces, given knowledge of the uniform phases.

I. INTRODUCTION

Recently we have developed a Ginzburg-Landau theory for the solid-liquid interfacial tension and interfacial width of the bcc elements.[1] This theory complements a number of other recent calculations of the solid-liquid interface, based on a density-functional formalism.[2–6] Our theory involves expanding the density functional in a power series, in which the individual coefficients are derived either from the properties of the pure liquid or from the freezing properties of the homogeneous system at melting. The theory gives surprisingly good agreement with experiment for the bcc elements Na and Fe, even though it *explicitly* involves only one order parameter, the amplitude of the [110] Fourier components of the density.

In this paper, we further test this theory by applying it to a broad range of other systems. We consider three in particular: the classical Wigner crystal, otherwise known as the one-component plasma (OCP); the quantum Wigner crystal, composed of interacting charged fermions; and the quantum Bose crystal ⁴He. We find that, to a first approximation, all can be treated by the method of Ref. 1, suitably generalized.

As formulated in,[1] our theory has only one principal order parameter, namely, the Fourier component of the density at some particular reciprocal lattice vector. Such a theory is not adequate to treat freezing of the uniform solid, which can be quantitatively analyzed only with the use of many reciprocal lattice vectors. The one-parameter interface theory avoids this problem by fitting the necessary coefficients to the freezing properties. Although this fitting procedure explicitly involves only one density coefficient, other coefficients (in particular, the liquid-solid density difference) are included implicitly via

perturbation theory. We find that the resulting expansion works surprisingly well for many kinds of interfaces between bcc solids and coexisting liquids.

The freezing transition is normally first order. Thus, if it is to be described by a Landau theory with only one order parameter, that Landau theory must include a nonzero third-order term. In Ref. 1, the order parameter was chosen to be the [110] Fourier density amplitudes, all of which must be equal by symmetry in the bcc solid. The [110] reciprocal lattice vectors have the property that groups of three such vectors can be chosen which sum to zero.[2] This leads to a nonvanishing third-order term in the Landau expansion, and hence a first-order freezing transition. Furthermore, the [110] reciprocal lattice vectors have nearly the same k value as the principal peak in the liquid structure factor.[3] The [110] vectors are therefore the softest of the "soft modes" in the liquid, and are the natural choice for an order parameter. In contrast, for fcc structures, the cubic term is absent if only the Fourier components of the smallest reciprocal lattice vector are chosen as order parameters. To obtain the observed first-order freezing transition in the fcc lattice, one must generally include at least two sets of Fourier components.[3,4]

The classical OCP can be treated directly within the formalism of Ref. 1 because it freezes into a bcc crystal.[7–14] The [110] amplitudes can also be used to treat the bcc Fermi and Bose crystal-melt interfaces. However, the classical Ginzburg-Landau theory cannot be directly used in these cases, because the quantum nature of the fluids requires the use of different linear response functions. We obtain the necessary response function from the Feynman theory[15] of liquid ⁴He.

We turn now to the body of the paper. The results for the OCP and the Fermi and the Bose crystal-melt inter-

face are presented in Secs. II, III, and IV. A brief discussion follows in Sec. V.

II. CLASSICAL ONE-COMPONENT PLASMA

A three-dimensional classical one-component plasma (OCP) is defined as a system of classical point ions, of charge Ze, where $-e$ is the charge of an electron, immersed in a uniform, rigid negative background of opposite charge. According to both Monte Carlo studies and analytic estimates,[7-14] the classical OCP undergoes a first-order freezing transition from a liquid to a body-centered-cubic (bcc) solid. The transition occurs at a value of the plasma parameter $\Gamma = Z^2 e^2 / (a k_B T)$ given by $\Gamma_f \sim 180$, where $a = (4\pi n_0/3)^{-1/3}$ is radius of a sphere containing exactly one ion and n_0 is the ionic number density. The entropy change per ion at this transition is estimated to be $\Delta \sim 0.82 k_B$. Since the neutralizing background of the OCP is rigid, charge neutrality leads to the constraint that the volume change on freezing is zero.

The interfacial tension of the OCP can easily be estimated using the density-functional approach of Ref. 1. In this approach, the grand free energy Ω is expressed as a functional of the singlet ionic number density $n(\mathbf{x})$,

$$\Omega = \Omega[n(\mathbf{x}); T] . \tag{1}$$

For a bulk solid or a bulk liquid, $n(\mathbf{x})$ can be expanded in terms of the Fourier components of the liquid density,

$$n(\mathbf{x}) = n_0 \left[1 + \eta + \sum_{\mathbf{K}_n \neq 0} u_n \exp(i\mathbf{K}_n \cdot \mathbf{x}) \right] , \tag{2}$$

where n_0 is the number density of the uniform liquid and $\mathbf{K}_n$ is a reciprocal lattice vector of the solid phase. The amplitudes η (for OCP $\eta = 0$) and u_n are constants in the uniform solid phase, and zero in the liquid phase. If there is a planar interface between solid and liquid perpendicular to the z axis, then the amplitudes η and u_n are z dependent and

$$n(\mathbf{x}) = n_0 \left[1 + \eta(z) + \sum_{\mathbf{K}_n \neq 0} u_n(z) \exp(i\mathbf{K}_n \cdot \mathbf{x}) \right] . \tag{3}$$

In order to calculate the solid-liquid interfacial tension of the OCP at melting, we assume that the dominant amplitudes at the transition are those corresponding to [110] reciprocal lattice vectors. The grand free energy difference per unit surface area, $\Delta \omega = \omega_{\text{sol}} - \omega_{\text{liq}}$, can then be expressed in the form of a Ginzburg-Landau expansion,

$$\Delta \omega = (n_0 k_B T/2) \int \left| a_2 u_{110}^2(z) - a_3 u_{110}^3(z) \right.$$
$$\left. + a_4' u_{110}^4(z) + b \left[\frac{du_{110}(z)}{dz} \right]^2 \right| dz . \tag{4}$$

As noted in Ref. 1, this procedure does *not* neglect coefficients other than the [110] set, but includes them implicitly via a kind of perturbation expansion. In particular, the density difference between solid and liquid, if

any, is not neglected. The form of the perturbation expansion is, however, such that only the [110] coefficients ultimately appear *explicitly* in the Ginzburg-Landau free energy.

The coefficients a_2, a_3, a_4', and b can be related to the properties of the bulk solid and of the liquid-solid transition. The result of this fitting procedure is

$$a_2 = 12/S(K_{110}) , \tag{5}$$

$$b = -\tfrac{1}{2} C''(K_{110}) \sum_{\hat{\mathbf{K}}_{110}} (\hat{\mathbf{K}}_{110} \cdot \hat{\mathbf{z}})^2 = -2C''(K_{110}) , \tag{6}$$

$$a_3 = (2 a_2 a_4')^{1/2} , \tag{7}$$

$$a_4' = (a_2/u_{110}^2) , \tag{8}$$

where $S(k)$ is the liquid structure factor, $C(k)$ is the direct correlation function defined by

$$S(k) = 1/[1 - C(k)] , \tag{9}$$

and $C''(k)$ is the second derivative $d^2 C(k)/dk^2$.

As noted in Ref. 1, Eqs. (5) and (6) are exact relations between a_2 and b and the linear response functions $S(k)$ and $C(k)$ of the homogeneous liquid. The relations (7) and (8), on the other hand, follow from the melting condition, $\Delta \omega = 0$ at the melting temperature, and from the extremal condition, $\partial \Delta \omega / \partial u_{110} = 0$, which must be satisfied by the free energy functional of the uniform solid at the melting temperature.

Since the linear response functions $S(k)$ and $C(k)$ of the liquid are readily estimated either from experiments or from various theories of uniform liquids, the coefficients a_2 and b are readily computed. Following the procedure of Ref. 1, we determine a_4' (or equivalently, u_{110}) by requiring that the expansion (4) give the correct heat of fusion at melting. This leads to the condition

$$\Delta = \tfrac{1}{2} T_m u_{110}^2 \left[\frac{da_2}{dT} \right]_{T = T_m} , \tag{10}$$

where Δ is the increase in entropy per ion on melting. Equations (8) and (10) together determine a_4' in terms of a_2 and its constant-volume temperature derivative $(da_2/dT)_{T=T_m}$, which can in turn, via Eq. (5), are related to the temperature derivative of the structure factor at melting, $[dS(K_{110})/dT]_{T=T_m}$.

Given the coefficients in the free-energy expansion, the corresponding surface tension and surface profile are obtained by minimizing the free-energy functional with respect to the amplitude $u_{110}(z)$. This minimization can be done analytically, with the results

$$\tau = (n_0 k_B T/6) u_{110}^2 (a_2 b)^{1/2} , \tag{11}$$

$$u_{110}(z) = u_{110}[1 + \tanh(\alpha z)] , \tag{12}$$

$$\alpha = [a_2/(4b)]^{1/2} . \tag{13}$$

Equations (5)–(13) give a complete description of the surface tension of any bcc elements, under the assumption of isotropy. By isotropy we mean that the coefficients u_{110} are taken to be equal for all members of

the [110] set of reciprocal lattice vectors, even within the surface region. The resulting surface tension is independent of surface orientation, by construction. Presumably, it can be viewed as a kind of average of τ over surface orientation.

This formalism can be applied in a straightforward way to the liquid-solid interface of the OCP, using as inputs the relevant results of Monte Carlo simulations.[7-10] From these simulations, one can extract the parameters $\Gamma_f \sim 180$; $S(K_{110}, \Gamma_f) \sim 2.97$; $C''(K_{110}, \Gamma_f) \sim -4.5a^2$;[2,9,10] $\Delta \sim 0.82 k_B$ per ion; and $(da_2/d\Gamma)|_{\Gamma = \Gamma_f} \sim 0.0108$. The resulting surface tension and surface width at freezing are

$$\tau_{OCP} = 1.12 \times 10^{-3} (Z^2 e^2/a^3) , \tag{14}$$

$$w_{OCP} = 6.6a , \tag{15}$$

where w_{OCP} is the 10-90 width of the interface profile (the distance over which the principal order parameter rises from 10% to 90% of its bulk solid value) and is related to the wave vector α of the hyperbolic tangent profile by

$$w_{OCP} = 2.2/\alpha . \tag{16}$$

To our knowledge, no computer simulations are available for the crystal-melt interface of the classical three-dimensional OCP. In order to provide a qualitative test, we compare these predictions with measurements and other calculations for Na, which is well described in terms of a classical OCP reference system.[16] The ionic sphere radius for Na at melting is $a = 2.14$ Å, which gives, when combined with Eq. (14), $\tau_{Na} = 26.3$ erg/cm^2, in reasonable agreement with the quoted experimental value $\tau_{expt.} \sim 20$ erg/cm^2.[17] The calculated width $w \sim 14$ Å. This value is about nine atomic layers and is comparable to the width calculated by computer simulation for the Lennard-Jones crystal-melt interface.[18]

Although we have calculated the interfacial tension for the OCP only in the isotropic approximation, we can crudely estimate the degree of anisotropy, by assuming it is comparable to that previously calculated for Na.[1] This analogy suggests an anisotropy of the order of 1% (Ref. 1) among the principal crystal faces.

III. CHARGED FERMION SYSTEM

The crystal-melt interfacial tension of a charged fermion system (that is, a Wigner crystal in equilibrium with its melt at 0 K) can, in principle, be calculated in a framework similar to Sec. II. However, some extension of this formalism is required to take into account the quantum nature of the fermion system. Moreover, the relevant correlation functions are very poorly known in the vicinity of the liquid-solid transition, which occurs at a much lower density than the metallic densities for which the electron-gas response functions are available. In this section, we outline the relevant formalism in such a way that the interfacial tension can be calculated easily, given the necessary correlation functions.

The fermion system at 0 K is described by the dimensionless coupling density parameter $r_s = (4\pi n_0/3)^{-1/3}/a_B$, where n_0 is the number density, $a_B = \hbar^2/(m_e e^2)$ is the Bohr radius of a fermion (which we

hereafter assume to be an electron), and m_e is the mass of an electron. The critical value, r_{sf}, at which the electron liquid freezes into a bcc Wigner crystal has been estimated by a number of groups, using both computer simulations and analytical approximations. The value of r_{sf} as estimated by these calculations ranges from 5 to 100. Presumably, the most reliable estimate would be that obtained from fermion Monte Carlo simulations,[19] which give $r_{sf} = 67$.

To calculate the surface tension of the quantum Wigner crystal-melt interface by the formalism of Sec. II, we must again take as order parameters the Fourier components of density corresponding to the first shell of reciprocal lattice vectors K_{110}. The excess grand free energy per unit surface area $\Delta \omega$ is then expanded as a power series in these order parameters. Assuming a planar crystal-melt interface normal to the z axis, we can conveniently write this expansion in the form

$$\Delta \omega = \lambda_1 \int dz \left| a_2 u_{110}^2(z) - a_3 u_{110}^3(z) + a_4' u_{110}^4(z) \right.$$
$$\left. + b \left[\frac{du_{110}(z)}{dz} \right]^2 \right| , \tag{17}$$

where $\lambda_1 = 2n_0^2/\lambda_0$ and $\lambda_0 = m_e k_F/(\pi \hbar)^2$, n_0 is the density of the electron plasma, and $k_F = (3\pi^2 n_0)^{1/3}$ is the Fermi wave vector of the electron plasma.

As in the classical OCP, the coefficients a_2 and b can be expressed in terms of the appropriate correlation functions of the uniform electron plasma,

$$a_2 = 12\bar{\chi}^{-1}(K_{110}) , \tag{18}$$

$$b = \frac{1}{2} [\bar{\chi}^{-1}(k)'']_{k = K_{110}} \sum_{\hat{K}_{110}} (\hat{K}_{110} \cdot \hat{z})^2 , \tag{19}$$

where $\bar{\chi}(k) = \chi(k)/\lambda_0$ is a dimensionless form of the density-density response function $\chi(k)$. These equations ultimately come from an expansion of the excess grand free energy $\Delta \Omega$ of an inhomogeneous system in powers of the density,

$$\Delta \Omega = \frac{1}{2} \int \int dx\, dx' G(x - x')[n(x) - n_0][n(x') - n_0] , \tag{20}$$

where the kernel $G(x)$ is related to $\chi(k)$ via

$$G(x) = \frac{1}{(2\pi)^3} \int dk \exp(i k \cdot x) \chi^{-1}(k) . \tag{21}$$

But whereas in the classical case $\chi(k)$ is related to the liquid structure via

$$\chi(k) = n_0 S(k)/k_B T , \tag{22}$$

$\chi(k)$ for the Fermi liquid is connected to the static dielectric function $\epsilon(k)$ via

$$\chi(k) = \frac{k^2}{4\pi^2 e^2} \left[1 - \frac{1}{\epsilon(k)} \right] , \tag{23}$$

and takes the form

$$\chi(k)=\frac{\chi_0(k)}{1+[1-G(k)]\chi_0(k)V_c(k)} \ , \tag{24}$$

where

$$V_c(k)=4\pi e^2/k^2 \ , \tag{25}$$

and

$$\chi_0(k)=\frac{m_e k_F}{\pi^2\hbar^2}\left[\frac{1}{2}+\frac{1-x^2}{4x}\ln\left|\frac{1+x}{1-x}\right|\right] \ . \tag{26}$$

Here $\chi_0(k)$ is the free-electron density response function and $x=k/2k_F$. The function $G(k)$, related to the irreducible polarization kernel, describes the effects of exchange and correlation on the static response of the electron gas.[20–22]

The remaining coefficients a_3 and a_4' in the Ginzburg-Landau expansion can be obtained by using melting and extremal conditions, as in Sec. II. The conditions again lead to Eqs. (7) and (8), which express a_3 and a_4' in terms of the single unknown u_{110}. Given u_{110}, a_2, and b, the solid-liquid interfacial tension of the Wigner crystal can then be expressed in a form identical to that of Sec. II:

$$\tau=\tfrac{1}{3}\lambda_1 u_{110}^2(a_2 b)^{1/2} \ . \tag{27}$$

The coefficients a_2 and b are now given by Eqs. (18) and (19). The Fourier component u_{110}^2 can be estimated in various ways. For example, one can employ a Lindemann melting criterion, as was done by Albers and Gubernatis,[23] to express u_{110} as a fraction of the nearest neighbor separation at melting. However, the other coefficients depend on the response function $\chi(k)$ at a very low density. Since this is poorly known, it is difficult to estimate the solid-liquid interfacial tension of a Wigner crystal at present. However, when a better response function becomes available, an estimate can be made very easily. Presumably, the resulting surface tension will be similar to that given in Eq. (14), with a different numerical coefficient.

IV. BOSON SYSTEM: He[4]

We turn next to a completely different type of solid-liquid interface, namely, that between a Bose fluid and its coexisting solid. The principal example in practice is solid bcc or hcp He[4] in equilibrium with its liquid along the melting curve in the P-T plane. The surface tension of this system has been measured along the melting curve by Gallet et al.[24] and by Balibar and Edwards.[25] These interfaces show a number of fascinating phenomena, such as the universal curvature of the crystal-melt interface in the vicinity of the roughening transition.[26] Our principal objective in this section is not to treat such subtle effects, but rather to show that the density-functional approach is at least qualitatively reasonable in boson systems as it is in classical and fermion crystals.

The details of the density functional are somewhat different for a Bose system than for a classical system. In terms of the Ginzburg-Landau form introduced in Sec. II, the difference arises in the coefficients a_2 and b: both involve integrals over an appropriate linear response

function $\chi(k)$. $\chi(k)$ appears in the density-functional formalism when the grand free energy of an inhomogeneous system is expanded about that of a homogeneous fluid. The result of this expansion, to second order in the density fluctuation, is same as Eq. (20) in Sec. III.

In the case of Bose liquids, as in Fermi liquids, $\chi(k)$ is not related to the structure factor by a classical relation such as Eq. (22). In order to treat the Bose crystal-melt interface at $T=0$, we estimate $\chi(k)$ within the Feynman approximation,[15] which is sufficiently accurate to give the roton minimum in the density fluctuation excitation spectrum at $T=0$,

$$\chi(k)=4n_0 MS(k)^2/(\hbar k)^2 \ , \tag{28}$$

where M is the atomic mass. As in Sec. III, it is convenient to introduce a dimensionless response function which takes the form $\bar\chi(k)=\chi(k)/\lambda_2$, where $\lambda_2=4n_0 M/(\hbar^2 K_{110}^2)$.

We now use the response function (28) to develop a Ginzburg-Landau approximation to the density functional for bcc He[4] at $T=0$. For bcc crystals, the order parameter corresponding to the smallest reciprocal lattice vector K_{110} is already sufficient to describe the first-order melting transition. To treat the nonuniform crystal-melt interface, we must allow the order parameters to vary in space. The free energy must now include a term involving the gradient of the order parameter. The appropriate expression for the excess grand free energy per unit surface area is (assuming a planar interface perpendicular to the z axis)

$$\Delta\omega[u_{110}(z)]=\frac{n_0\hbar^2 K_{110}^2}{8M}$$
$$\times\int dz\left[a_2 u_{110}^2(z)-a_3 u_{110}^3(z)\right.$$
$$\left.+a_4' u_{110}^4(z)+b\left[\frac{du_{110}(z)}{dz}\right]^2\right] \ , \tag{29}$$

where once again a_2, a_3, a_4', and b are coefficients to be determined.

The coefficients a_2 and b can now be obtained by use of Eqs. (18) and (19) in Sec. III, using a derivation analogous to that used in finding these coefficients for the classical case. The results are (assuming the Feynman form for the response function)

$$a_2=12/\bar\chi(K_{110})=12/S^2(K_{110}) \ , \tag{30}$$

and

$$b=\frac{1}{S^2(K_{110})}\sum_{\hat{\mathbf{K}}_{110}}(\hat{\mathbf{K}}_{110}\cdot\hat{\mathbf{z}})^2$$
$$\times\left[\frac{1}{K_{110}^2}-\frac{4S'(K_{110})}{K_{110}S(K_{110})}-\frac{S''(K_{110})}{S(K_{110})}\right.$$
$$\left.+\frac{3S'(K_{110})^2}{S(K_{110})^2}\right] \ . \tag{31}$$

As in Sec. II, the coefficients a_3 and a_4' can be expressed in terms of the coefficient a_2 and the bulk solid order parameter u_{110}, using the melting condition and the requirement that the free energy be an extremum with respect to the order parameter. The expressions for these coefficients are identical to Eqs. (7) and (8). Now, as has been shown by Ramakrishnan and Yussouff,[3] the bulk order parameter u_{110} is related to the Debye-Waller factor $e^{-2W_{110}}$ (Ref. 3) through the relation $\bar{u}_{110}^2 = e^{-2W_{110}}$, where $\bar{u}_{110} = u_{110}/(1+\eta)$ and η is the fractional density increase on freezing. For the OCP $\eta = 0$ (a consequence of the Coulomb forces in these materials), and thus $\bar{u}_{110} = u_{110}$. In the Bose system, by contrast, the forces are short range and these two parameters are not equal. At $T = 0$, the Debye-Waller factor is due entirely to the zero-point vibrations of the quantum crystal, and may be approximated by the expression

$$W = 3\hbar^2 K_{110}^2/(8Mk_B\Theta_D) , \qquad (32)$$

where Θ_D is the Debye temperature of the solid, and for bcc helium we use the estimate $\Theta_D = 21.4K$, as extrapolated to $T = 0$ by Edwards and Pandorf.[27]

The interfacial tension for a planar surface can be obtained by minimizing the excess grand free energy functional as in Secs. II and III. Minimizing the relevant functional, expression (29), gives

$$\tau = n_0\hbar^2 K_{110}^2 u_{110}^2 (a_2b)^{1/2}(24M) . \qquad (33)$$

We now apply this phenomenological theory to estimate the solid-superfluid interfacial tension of bcc ^{4}He at $T = 0$. The calculation requires the structure factor of liquid He^4 at $T = 0$ and at a pressure of about 25 atm, comparable to the freezing pressure; we take this function from Green's function Monte Carlo calculations,[28] which agrees very well with x-ray[29] and neutron diffraction[30,31] studies at 1 atm pressure. Our results for the input coefficients (namely, a_2, b, and the value of u_{110} in the solid phase) are shown in Table I, as are both the "experimental" and calculated bcc surface tensions at $T = 0$. Since the bcc solid is metastable at $T = 0$, the experimental results must be extrapolated from measurements of the surface tension and its temperature derivative in the bcc phase in the range $T = 1.46-1.76$ K;[25] these measurements are given in Table I. Agreement between theory and linearly extrapolated experiment is reasonable, especially considering the approximations required to obtain both theoretical and experimental estimates. The agreement between theory and experiment is actually better than that shown, because the linearly extrapolated "experimental" surface tension is certainly too large. Indeed, the derivative $-S_{sur} = d\tau/dT = 0$ at $T = 0$, since the surface entropy S_{sur} must vanish at $T = 0$. Thus the extrapolated experimental curve must reach $T = 0$ with horizontal slope.

V. DISCUSSION

We have presented a formalism which permits a simple calculation of the crystal-melt interfacial tension for three systems: the classical OCP, the quantum electronic system at zero temperature (quantum Wigner crystal), and the boson system (^{4}He). All three calculations represent an application of a density-functional formalism in a Ginzburg-Landau approximation. In all three cases, we have also made an isotropic approximation, so that the calculations involve an orientational average of information about the interface. The formalism could readily be extended to calculate orientation-dependent properties, but for these, it would certainly be necessary to include more order parameters in the expansion of the free energy.

Our Bose calculations agree reasonably well with an extrapolation of experimental data for ^{4}He to 0 K. Likewise, the OCP calculations are in good agreement with experimental data for Na, as inferred from nucleation theory applied to supercooling experiments. This comparison is germane because Na, of all the simple metals, is the best described by the OCP model. The fermion formalism is more difficult to apply at present because the necessary electron-gas response functions are not available at the densities of the Wigner transition.

Our results for both the OCP and ^{4}He appear to be roughly described by a simple rule of thumb of the type first suggested by Turnbull.[32] For the OCP, Turnbull's rule suggests that the liquid-solid interfacial tension should be approximately the heat of fusion per surface atom. The heat of fusion is $T_m\Delta$, where T_m is the melting temperature and Δ is the entropy of fusion. If we consider the [100] surface of the solid OCP, and consider as surface atoms only those atoms in the outermost surface layer, this estimate gives $\tau \approx 1.15 \times 10^{-3} Z^2e^2/a$, very close to our result (14). This argument cannot be used directly for ^{4}He at 0 K, because, of course, $\Delta = 0$ at 0

TABLE I. Input parameters and calculated results for bcc ^{4}He. n_0 is the number density of liquid ^{4}He near freezing at pressure about 25 atm; $\eta = (n_s - n_0)/n_0$ is the fractional density increase on melting; coefficients a_2, b, and u_{110}, all as defined in the text; the experimental surface tension τ_{expt} is the linearly extrapolated result as explained in the text; and w is the 10-90 width of the interface.

n_0 (Å^{-3})	η	a_2	b (Å^2)	u_{110}	τ_{theor} (erg/cm^2)	τ_{expt} (erg/cm^2)	w (Å)
0.0258	0.0853	3.7	~10	0.41	0.085	0.21 ($T = 0$ K)	7.2
						0.112[a] ($T = 1.46$ K)	
						0.088[a] ($T = 1.76$ K)	

[a]Reference 25.

K. A reasonable alternative is $\tau \approx P \Delta v n_{sur}$, where P is the coexistence pressure, Δv is the change in atomic volume at freezing, and n_{sur} is the number of surface atoms per unit area. This formula gives $\tau \approx 0.04-0.07$ erg/cm^2, somewhat lower than the extrapolated experimental value but in the same range.

The reasonable agreement between our Ginzburg-Landau calculation and an extrapolation of the experimental ^{4}He surface tensions indicates that our extremely simple phenomenological theory is also plausible for quantum Bose systems. Thus the density-functional approach, suitably generalized to include more order parameters, seems to be among the most promising presently available for both classical and quantum systems. With the inclusion of more order parameters and better response functions, it may be possible in the future to obtain truly quantitative agreement with experiment. It may also be useful for other types of systems not considered in this paper, such as the interface between solid bcc ^{3}He and its liquid at $T = 0$.

ACKNOWLEDGMENTS

[1]W. H. Shih, Z. Q. Wang, X. C. Zeng, and D. Stroud, Phys. Rev. A **35**, 2611 (1987).

[2]S. Alexander and J. McTague, Phys. Rev. Lett. **41**, 702 (1978).

[3]T. V. Ramakrishnan and M. Yussouff, Phys. Rev. B **19**, 2275 (1979).

[4]D. W. Oxtoby and A. D. J. Haymet, J. Chem. Phys. **76**, 6262 (1982).

[5]C. Ebner, W. F. Saam, and D. Stroud, Phys. Rev. A **14**, 2264 (1976).

[6]W. A. Curtin and N. W. Ashcroft, Phys. Rev. Lett. **56**, 2775 (1986); W. A. Curtin, *ibid*. **59**, 1228 (1987).

[7]J. P. Hansen, Phys. Rev. A **8**, 3096 (1973).

[8]E. L. Pollock and J. P. Hansen, Phys. Rev. A **8**, 3110 (1973).

[9]W. L. Slattery, G. D. Doolen, and H. E. Dewitt, Phys. Rev. A **26**, 2255 (1982); **21**, 2087 (1980).

[10]F. J. Rogers, D. A. Young, H. E. Dewitt, and M. Ross, Phys. Rev. A **28**, 2990 (1983).

[11]A. D. J. Haymet, Phys. Rev. Lett. **52**, 1013 (1984).

[12]B. Bagchi, C. Cerjan, U. Mohanty, and S. A. Rice, Phys. Rev. B **29**, 2857 (1984).

[13]M. Rovere and M. P. Tosi, J. Phys. **18**, 3445 (1985).

[14]N. H. March and M. P. Tosi, Phys. Chem. Liq. **14**, 303 (1985).

[15]R. P. Feynman and M. Cohen, Phys. Rev. **102**, 1202 (1956).

[16]K. K. Mon, R. Gann, and D. Stroud, Phys. Rev. A **24**, 2145

[17]J. W. Taylor, Philos. Mag. **46**, 857 (1955).

[18]J. Q. Broughton and G. H. Gilmer, J. Chem. Phys. **84**, 5749 (1986).

[19]D. Ceperley, Phys. Rev. B **18**, 3126 (1978).

[20]J. Hubbard, Proc. R. Soc. London, Ser. A **243**, 336 (1958).

[21]K. Utsumi and S. Ichimaru, Phys. Rev. B **22**, 5203 (1980).

[22]S. Ichimaru and K. Utsumi, Phys. Rev. B **24**, 7385 (1981).

[23]R. C. Albers and J. E. Gubernatis, Phys. Rev. B **23**, 2782 (1981).

[24]F. Gallet, P. E. Wolf, and S. Balibar, Phys. Rev. Lett. **52**, 2253 (1984).

[25]S. Balibar, D. O. Edwards, and C. Laroche, Phys. Rev. Lett. **42**, 782 (1979).

[26]C. Jayaprakash, W. F. Saam, and S. Teitel, Phys. Rev. Lett. **50**, 2017 (1983).

[27]D. O. Edwards and R. C. Pandorf, Phys. Rev. **144**, 143 (1966).

[28]D. M. Ceperley, G. V. Chester, M. H. Kalos, and P. A. Whitlock, J. Phys. (Paris), Colloq. **C6**, 1298 (1978).

[29]F. K. Achter and L. Meyer, Phys. Rev. **188**, 291 (1969).

[30]D. G. Henshaw, Phys. Rev. **119**, 9 (1960).

[31]B. Mozer, L. A. De Graaf, and B. Le Neindre, Phys. Rev. A **9**, 448 (1974).

[32]D. Turnbull, J. Appl. Phys. **21**, 1022 (1950).

(1981).