Pressure and density of vacancies in solid $^4\text{He}$

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Crystals of $^4\text{He}$ contain vacancies that move around by a quantum mechanical hopping process. The density and pressure of these vacancies can be experimentally studied. The accuracy of the experiments is high enough to detect the effect of the Bose statistics of the vacancies. In this paper we examine the effect of the hard-core repulsion between the vacancies, which should also have a measurable effect on their behaviour. We set up a virial expansion for a lattice gas of hard-core particles, and calculate the second virial coefficient. It turns out that the vacancies behave as ideal Bose particles at low temperatures, but that the hard-core interaction makes them behave more and more like fermions as the temperature increases.

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I. INTRODUCTION

The thermodynamic behaviour of vacancies in solid $^4\text{He}$ is an interesting experimental and theoretical problem [1]. Vacancies in Helium are more mobile than in any other solid. At the temperatures where solid Helium exists the motion of vacancies requires a quantum mechanical description. They hop from site to site with a certain rate $\nu_v$, leading to a band of states $\varepsilon(\vec{k})$, much like the electron motion in the tight binding approximation. For $^4\text{He}$ the vacancies are obviously bosons, since the creation operator for a vacancy is the annihilation operator for a $^4\text{He}$ particle, which is a boson [2]. The fact that two vacancies cannot occupy the same lattice site has to be incorporated as a hard-core potential for the hopping bosons. So the simplest model for vacancies in $^4\text{He}$ is that of a gas of hard-core bosons on a lattice. In reality the strain fields around the vacancies produce a more complicated interaction between them than the simple on-site exclusion. However, we consider the hard-core boson approximation as a sufficiently realistic description of the vacancy motion to leave out these further refinements, in order not to complicate the model too much. The hard-core bosons on a lattice are known to be equivalent to a spin-$\frac{1}{2}$ quantum system with an interaction of the XY type [3]. It is however not this analogy that is exploited in this paper. The reason is that the vacancy system is in practical circumstances always extremely dilute, which corresponds in the spin analogy to a system in an extremely large external magnetic field.
In this limit the spin analogy is of little use and in our opinion less transparent than the particle language.

Experiments show that the percentage of vacancies in a crystal is at most of the order of 1% \[4\], at least at temperatures of the order of 1K, where the experiments take place. As a first approximation the vacancies thus behave as an ideal gas. However, present day experiments are sufficiently accurate that effects of Bose statistics can be detected. It is one of our points that then also effects of the hard-core interaction become detectable.

In this paper we present a systematic analysis of the vacancies using \(\exp(-\Delta/k_BT)\) as a small parameter, \(\Delta\) being the excitation energy, or band gap, required to create a vacancy. This is equivalent to a virial expansion \([4]\) for the quantum lattice gas, and we work out the properties in detail up to the second virial coefficient. A general formula for the second virial coefficient of the hard-core Bose lattice gas is derived and evaluated for hypercubic lattices in one, two, and three dimensions. The case of the hcp lattice, which applies to real solid \(^4\)He, will be treated elsewhere.

II. THE HARD-CORE BOSE LATTICE GAS

The vacancies are represented by Bose creation and annihilation operators \(\hat{b}_i^\dagger, \hat{b}_i\) obeying the usual Bose commutation relations. The Hamiltonian for the vacancies is given by

\[
\mathcal{H} = -t \sum_{\langle i,j \rangle} (\hat{b}_i^\dagger \hat{b}_j + \hat{b}_j^\dagger \hat{b}_i) + (\Delta + ct) \sum_i \hat{b}_i^\dagger \hat{b}_i + \frac{U}{2} \sum_i \hat{b}_i^\dagger \hat{b}_i \hat{b}_i \hat{b}_i.
\]

(2.1)

Here the transfer integral \(t\) is equal to \(\hbar \nu_v\), and the hops take place between all pairs of nearest neighbours \(\langle i, j \rangle\) on the lattice. The coordination number of the lattice is \(c\), and \(\Delta\) is the energy required to create a vacancy. It functions as minus the chemical potential for the vacancies. The last term represents the vacancy-vacancy repulsion. It could be omitted in favour of a change to on-site Fermi commutation relations for the \(\hat{b}_i^{(t)}\), but we prefer to work with standard commutation relations and a potential \(U\) which penalizes the simultaneous occurrence of two or more vacancies at the same site. We will let \(U \to \infty\), or \(U\) is much larger than any other energy in the problem.

Without the potential term we have an ideal Bose lattice gas, the Hamiltonian of which can be diagonalized by the canonical transformation

\[
\hat{b}(\vec{k}) = \frac{1}{\sqrt{N}} \sum_j \hat{b}_j e^{i\vec{k} \cdot \vec{r}_j},
\]

(2.2)

\[
\hat{b}_j = \frac{1}{\sqrt{N}} \sum_{\vec{k}} \hat{b}(\vec{k}) e^{-i\vec{k} \cdot \vec{r}_j},
\]
where $N$ is the number of sites in the system. We use periodic boundary conditions, and the wave vectors $\vec{k}$ are restricted to the first Brillouin zone of the lattice.

Inserting (2.1) into the first two terms of (2.2) we obtain the unperturbed Hamiltonian

$$
H_0 = \sum_k [\Delta + \varepsilon(\vec{k})] b^\dagger(\vec{k}) b(\vec{k}),
$$

(2.3)

where $\varepsilon(\vec{k})$ is given by

$$
\varepsilon(\vec{k}) = t \sum_\delta (1 - \cos \vec{k} \cdot \vec{r}_\delta),
$$

(2.4)

and $\vec{r}_\delta$ is the set of the $c$ nearest neighbour positions with respect to a centrally chosen site. $\Delta$ is the gap of the energy band, since we have $\varepsilon(\vec{0}) = 0$ as lowest energy in the center of the Brillouin zone. One sees that $t$ is a measure for the bandwidth, which for a $d$-dimensional hypercubic lattice is $w = 4dt$.

The interaction is written in terms of $b^\dagger(\vec{k})$ as

$$
V = \frac{U}{2N} \sum_{\vec{k}, \vec{G}} \delta_{\vec{k}_1+\vec{k}_2+\vec{k}_3+\vec{k}_4+\vec{G}} b^\dagger(\vec{k}_1) b^\dagger(\vec{k}_2) b(\vec{k}_3) b(\vec{k}_4),
$$

(2.5)

where $\vec{G}$ is a vector of the reciprocal lattice. The matrix elements of the interaction have no other structure than the conservation of the total incoming and outgoing momentum (up to a reciprocal lattice vector), a feature which is of great advantage in solving the two-particle problem.

III. THE VIRIAL EXPANSION

The grand partition function of the vacancy system is given by

$$
\Xi = Tr \ e^{-\beta H},
$$

(3.1)

with $H$ given by (2.2), $\beta = 1/k_B T$, and $Tr$ stands for the trace over all symmetrized states. As $H$ conserves the number of vacancies, $\Xi$ can be expanded as

$$
\Xi = \sum_{n=0}^{\infty} Z_n e^{-n\beta \Delta},
$$

(3.2)

where $Z_n$ is the canonical partition sum for $n$ vacancies excluding the contribution from the gap $\Delta$. Of course, $Z_0 = 1$, and

$$
Z_1 = Tr_1 e^{-\beta (H_0 - \Delta)} = \sum_{\vec{k}} e^{-\beta \varepsilon(\vec{k})},
$$

(3.3)

because for one vacancy no hard-core effects enter. $Tr_n$ is the trace over $n$-vacancy states.
For ln $\Xi$ we may deduce from (3.2)

$$\beta p N v_0 = \ln \Xi = N \sum_{\ell=1}^{\infty} b_\ell e^{-\ell \beta \Delta}, \quad (3.4)$$

where $v_0$ is the volume of the unit cell, $p$ is the pressure of the vacancies, and the $b_\ell$ are the fugacity expansion coefficients. The first of these, $b_1$, reads

$$b_1(\beta) = \frac{1}{N} Z_1 = \frac{v_0}{(2\pi)^d} \int_{BZ} d\vec{k} e^{-\beta \varepsilon(\vec{k})}, \quad (3.5)$$

where we have replaced the sum over $\vec{k}$ by an integral over the Brillouin zone. In contrast to the continuum ideal gas, $b_1$ is not simply related to a thermal wavelength $\lambda$ as

$$b_1 = v_0/\lambda^d. \quad (3.6)$$

A formula of this type only results when the temperature is so low that $\varepsilon(\vec{k})$ may be replaced by its low-momentum behaviour

$$\varepsilon(\vec{k}) \approx \frac{t}{2} \sum_{\delta} (\vec{k} \cdot \vec{r}_\delta)^2 = \frac{\hbar^2 k^2}{2m^*}, \quad (3.7)$$

defining an effective mass $m^*$ for the vacancies. For $d$-dimensional hypercubic lattices we find

$$m^* = \frac{\hbar^2}{2ta^2}, \quad (3.8)$$

where $a$ is the lattice constant. Using (3.7) in (3.5) and extending the $\vec{k}$-integral beyond the Brillouin zone to infinity (as is allowed for large $\beta$ or small $T$) one finds (3.6) with

$$\lambda^2 = \frac{\hbar^2}{2\pi m^* k_B T}. \quad (3.9)$$

The second term $b_2$ in (3.4) is our main concern in this paper. We find from (3.2) and (3.4)

$$b_2 = \frac{1}{N} \left( Z_2 - \frac{Z_2^2}{2} \right) = \frac{1}{N} \left\{ \text{Tr}_2 e^{-\beta (H - 2\Delta)} - \frac{1}{2} \left( \text{Tr}_1 e^{-\beta (H_0 - \Delta)} \right)^2 \right\}. \quad (3.10)$$

We rewrite this expression by adding and subtracting the contribution of the non-interacting two-vacancy system. So we define

$$b_2^{\text{int}} = \frac{1}{N} \text{Tr}_2 \left[ e^{-\beta (H - 2\Delta)} - e^{-\beta (H_0 - 2\Delta)} \right] \quad (3.11)$$
as the contribution of the hard-core interaction, and

$$b_2^0 = \frac{1}{N} \left[ \text{Tr}_2 e^{-\beta (H_0 - 2\Delta)} - \frac{1}{2} \left( \text{Tr}_1 e^{-\beta (H_0 - \Delta)} \right)^2 \right]. \quad (3.12)$$
as the effect of the Bose statistics of the vacancies. The combination

\[ b_2 = b_2^0 + b_2^{\text{int}} \]

(3.13)
yields the total effect.

The statistical effects are trivial to calculate, as the unperturbed grand partition function is given by

\[ \ln \Xi^0 = -\sum_k \ln \left(1 - e^{-\beta(\varepsilon(k) + \Delta)}\right) = N \sum_{\ell=1}^\infty b_\ell^0 e^{-\ell\beta\Delta}, \]

(3.14)
such that

\[ b_\ell^0 = \frac{1}{N} \sum_k e^{-\ell\beta\varepsilon(k)} = \frac{1}{\ell} b_1(\beta\ell), \]

(3.15)
with \( b_1(\beta) \) given by (3.5). Thus it suffices to focus our attention on the calculation of \( b_2^{\text{int}} \). As a general observation we note that the two contributions in (3.13) will have opposite signs. To see this more clearly, we go over to a series in the density \( n \) of the vacancies,

\[ n = \frac{1}{nv_0} \sum_i b_i^\dagger b_i = -\partial p/\partial \Delta = \frac{1}{v_0} \sum_{\ell=1}^\infty \ell b_\ell e^{-\ell\beta\Delta}. \]

(3.16)
Eliminating \( e^{-\beta\Delta} \) from (3.4) and (3.16) we obtain a virial expansion for the pressure

\[ \beta p = n - (b_2/b_1^2)v_0 n^2 + \ldots. \]

(3.17)
So at fixed density \( n \) the statistical effects lower the pressure as \( b_2^0 \) is positive according to (3.15). The hard-core repulsion can only increase the pressure, so \( b_2^{\text{int}} \) must be negative.

**IV. THE SECOND VIRIAL COEFFICIENT**

The second virial coefficient (3.11) is evaluated as

\[ b_2^{\text{int}} = \frac{1}{N} \int dE \left[ \rho_2(E) - \rho_2^0(E) \right] e^{-\beta(E - 2\Delta)}, \]

(4.1)
where the level densities \( \rho_2(E) \) are given by

\[ \begin{cases} 
\rho_2(E) = \text{Tr}_2 \delta(E - \mathcal{H}) \\
\rho_2^0(E) = \text{Tr}_2 \delta(E - \mathcal{H}_0).
\end{cases} \]

(4.2)
The level densities are obtained from the formula

\[ G_+(E) = \frac{1}{E + i\epsilon - \mathcal{H}} = \mathcal{P} \frac{1}{E - \mathcal{H}} - i\pi \delta(E - \mathcal{H}), \]

(4.3)
which transfers the problem to the determination of the Green’s function, for which the general equation holds.
\[ \mathcal{G}(z) = \mathcal{G}_0(z) + \mathcal{G}_0(z)V\mathcal{G}(z), \]  
(4.4)

with \( z \) a complex number. The density \( \rho(E) \) is obtained from \( \mathcal{G} \) as

\[ \rho(E) = -\frac{1}{\pi} \text{Im} \text{Tr} \mathcal{G}_+(E). \]  
(4.5)

The states of the unperturbed 2-vacancy Hamiltonian are denoted by two wavenumbers \( \vec{k}_1 \) and \( \vec{k}_2 \) with the property

\[ \mathcal{H}_0|\vec{k}_1 \vec{k}_2\rangle = E_0(\vec{k}_1, \vec{k}_2)|\vec{k}_1 \vec{k}_2\rangle \]
\[ = \left( 2\Delta + \varepsilon(\vec{k}_1) + \varepsilon(\vec{k}_2) \right)|\vec{k}_1 \vec{k}_2\rangle. \]  
(4.6)

The matrix elements of \( V \) are obtained from (2.5)

\[ \langle \vec{k}_1 \vec{k}_2|V|\vec{k}_1' \vec{k}_2'\rangle = \frac{2U}{N} \sum_{\vec{G}} \delta_{\vec{k}_1+\vec{k}_2, \vec{k}_1'+\vec{k}_2'+\vec{G}}, \]  
(4.7)

which shows that a representation in center of mass and relative coordinates will be advantageous. Thus we introduce

\[ \vec{K} = \vec{k}_1 + \vec{k}_2, \quad \vec{k} = (\vec{k}_1 - \vec{k}_2)/2. \]  
(4.8)

The sum over \( \vec{G} \) is eliminated by choosing the Brillouin zone in such a way that no two points in it have values of \( \vec{K} \) differing by a reciprocal lattice vector, so that only the term with \( \vec{G} = \vec{0} \) contributes. The matrix element of \( V \) is then diagonal in \( \vec{K} \). From now on we will assume that the Brillouin zone has been chosen in such a way, and drop the reference to \( \vec{G} \). Since the total momentum \( \vec{K} \) is conserved by \( \mathcal{H}_0 \) and \( \mathcal{V} \), \( \mathcal{G} \) becomes diagonal in it. So, writing (4.4) in the \( \vec{K}, \vec{k} \) representation, we have

\[ \langle \vec{K} \vec{k}|\mathcal{G}(z)|\vec{K} \vec{k}'\rangle = \frac{1}{z - E_0(\vec{K}, \vec{k})} \left\{ \delta_{\vec{k}, \vec{k}'} ight. \]
\[ + \frac{2U}{N} \sum_{\vec{k}''} \langle \vec{k}'|\mathcal{G}(z)|\vec{k}''\rangle \left. \right\}. \]  
(4.9)

The simplifying feature of (4.9) is that the general matrix element of \( \mathcal{G} \) couples only to the total sum over the first entry of the matrix elements. For the latter we obtain an expression by summing (4.9) over \( \vec{k} \)

\[ \sum_{\vec{k}} \langle \vec{K} \vec{k}|\mathcal{G}(z)|\vec{K} \vec{k}'\rangle = \frac{1}{z - E_0(\vec{K}, \vec{k}')} \]
\[ + 2U \mathcal{R}(z, \vec{K}) \sum_{\vec{k}''} \langle \vec{k}'|\mathcal{G}(z)|\vec{k}''\rangle, \]  
(4.10)

with \( \mathcal{R}(z, \vec{K}) \) given by
\[
\mathcal{R}(z, \vec{K}) = \frac{1}{N} \sum_{\vec{k}} \frac{1}{z - E_0(\vec{K}, \vec{k})}.
\] (4.11)

Now (4.10) is an algebraic equation for the quantity in the left hand side. Using the solution of this equation in (4.9) one finds
\[
\langle \vec{K} \vec{k} \mathcal{G}(z) | \vec{K} \vec{k}' \rangle = \frac{1}{z - E_0(\vec{K}, \vec{k})} \left\{ \delta_{\vec{k}, \vec{k}'} + \frac{2U/N}{1 - 2U \mathcal{R}(z, \vec{K})} \frac{1}{z - E_0(\vec{K}, \vec{k}')} \right\}.
\] (4.12)

Note that the off-diagonal elements are of order \(N^{-1}\) while the diagonal elements are of order 1. With (4.12) we can calculate the level density from
\[
\text{Tr} \mathcal{G}(z) = \sum_{\vec{K}, \vec{k}} \langle \vec{K} \vec{k} \mathcal{G}(z) | \vec{K} \vec{k} \rangle,
\] (4.13)
leading to the compact expression
\[
\text{Tr} \mathcal{G}(z) = \text{Tr} \mathcal{G}_0(z) + \frac{\partial}{\partial z} \sum_{\vec{K}} \ln \left[ 1 - 2U \mathcal{R}(z, \vec{K}) \right],
\] (4.14)
which in turn yields for the difference \(\rho_2 - \rho_2^0\)
\[
\rho_2(E) - \rho_2^0(E) = -\frac{1}{\pi} \text{Im} \left[ \frac{\partial}{\partial z} \sum_{\vec{K}} \ln \left( 1 - 2U \mathcal{R}(z, \vec{K}) \right) \right] \bigg|_{z=E+i\epsilon}.
\] (4.15)

This expression holds generally for any on-site repulsion \(U\). We may let \(U \to \infty\), by which it will disappear from the formula as the \(U\) term under the logarithm starts to dominate the argument for any \(z\) and \(\vec{K}\). Omitting the 1 in the argument of the logarithm, the term \(\ln(-2U)\) drops out after differentiation with respect to \(z\). So for (4.15) we have in the limit \(U \to \infty\) the equivalent expression
\[
\rho_2(E) - \rho_2^0(E) = -\frac{1}{\pi} \text{Im} \left[ \frac{\partial}{\partial z} \sum_{\vec{K}} \ln \mathcal{R}(z, \vec{K}) \right] \bigg|_{z=E+i\epsilon}.
\] (4.16)

Hereby the problem is essentially reduced to the evaluation of \(\mathcal{R}(z, \vec{K})\) given by (4.11).

A few comments are in order about this expression. For bosons the state \(|\vec{k}_1 \vec{k}_2\rangle\) is the same as \(|\vec{k}_2 \vec{k}_1\rangle\). So the relative momenta \(\vec{k}\) and \(-\vec{k}\) should be identified with each other. Both \(\vec{k}_1\) and \(\vec{k}_2\) run through a Brillouin zone.
appropriate for the structure of the lattice, and this in principle defines the ranges of $\vec{K}$ and $\vec{k}$. But as was mentioned before, we have chosen the Brillouin zone such that there are no points whose values of $\vec{K}$ differ by a reciprocal lattice vector.

From the definition (4.11) of $R(z, \vec{K})$ and the expression (4.6) for $E_0(\vec{K}, \vec{k})$ one sees that $2\Delta$ occurs as a shift in the energy variable and the $\rho_2$ are functions of $E - 2\Delta$. Taking $E - 2\Delta$ as an integration variable in (4.1) one finds

$$b_2^{\text{int}} = \frac{1}{N} \int dE' \left[ \rho_2(E') - \rho_2^0(E') \right] e^{-\beta E'}, \quad (4.17)$$

with

$$\rho_2(E') - \rho_2^0(E') = -\frac{1}{\pi} \text{Im} \left[ \frac{\partial}{\partial z} \sum_{\vec{K}} \ln R'(z, \vec{K}) \right]_{z=E'+i\epsilon}, \quad (4.18)$$

and $R'(z, \vec{K})$ given by

$$R'(z, \vec{K}) = \frac{1}{N} \sum_{\vec{k}} \frac{1}{z - \varepsilon(\vec{K}/2 + \vec{k}) - \varepsilon(\vec{K}/2 - \vec{k})}. \quad (4.19)$$

In this formula $\Delta$ is eliminated, as it should be. Using the fact that $\text{Im} \ln z = \arg z$, we can further rewrite (4.18)

$$\rho_2(E') - \rho_2^0(E') = -\frac{N}{\pi} \frac{\partial}{\partial z} F(z) \big|_{z=E'+i\epsilon}, \quad (4.20)$$

with

$$F(z) = \frac{1}{N} \sum_{\vec{K}} \arg R'(z, \vec{K}). \quad (4.21)$$

From (4.18) and (4.19) one sees that only $E'$ values occur in (4.17) which lead to complex values of $R'(z, \vec{K})$. These occur when $z = E'+i\epsilon$ is a pole in the $\vec{k}$-integration in (4.19). Thus the combined bandwidth of $\varepsilon(\vec{K}/2 + \vec{k}) + \varepsilon(\vec{K}/2 - \vec{k})$ determines the range of $E'$ values. This is twice the bandwidth of $\varepsilon(\vec{k})$ just as in the ideal Bose contribution (3.15).

V. THE ONE-DIMENSIONAL CASE

We interrupt the general discussion for the treatment of the one-dimensional case of (4.17)-(4.21), as this case is interesting, completely analyzable, and elucidating for the structure of the functions occurring in (4.17)-(4.21).

The $d = 1$ band structure is given by (for a lattice constant $a = 1$)

$$\varepsilon(k) = 2t(1 - \cos k), \quad (5.1)$$

which gives for $b_1(\beta)$ the expression
\[ b_1(\beta) = \frac{1}{2\pi} \int_{-\pi}^{\pi} dk \, e^{-2\beta t(1-\cos k)} = e^{-2\beta t} I_0(2\beta t), \quad (5.2) \]

where \( I_0(x) \) is a modified Bessel function of the first kind.

For the evaluation of \( R(z, K) \) we rearrange the Brillouin zone in such a way that its boundaries in \( K, k \) space are convenient, and also such that no reciprocal lattice vector \( G \) enters into the problem. In figure 3 we have divided the original Brillouin zone \(-\pi < k_1 < \pi , -\pi < k_2 < \pi \) into four domains, I, II, III, and IV. Domains I and IV, as well as II and III, refer to the same states, as they are obtained from each other through interchanging \( k_1 \leftrightarrow k_2 \). So it suffices to take one of each, say I and III.

Now III is equivalent to III', which follows from III by shifting \( k_1 \) over \( 2\pi \). The combined domain I and III' is given in \( K, k \) space by \( 0 < K < 2\pi, 0 < k < \pi \).

Using this parameter space one can write (4.19) explicitly as (dropping the primes)

\[ R(z, K) = \frac{1}{2\pi} \int_{0}^{\pi} dk \, \frac{1}{z - 4t(1 - \cos \frac{K}{2}\cos k)}. \quad (5.3) \]

For convenience we put \( z = 4t(\zeta + 1 + i\epsilon), \) and have

\[ R(\zeta, K) = \frac{1}{8\pi t} \mathcal{P} \int_{0}^{\pi} dk \, \frac{1}{\zeta + i\epsilon + \cos \frac{K}{2}\cos k} \]

\[ - \frac{i}{8t} \int_{0}^{\pi} dk \, \delta(\zeta + \cos \frac{K}{2}\cos k). \quad (5.5) \]

Two cases must be distinguished: for \(|\zeta| > |\cos(K/2)|\), the imaginary part of \( R \) is zero, and for \(|\zeta| < |\cos(K/2)|\) its real part is zero. The result is

\[ R(\zeta, K) = \begin{cases} 
\frac{1}{8t} \frac{\text{sgn}(\zeta)}{\sqrt{\zeta^2 - \cos^2 \frac{K}{2}}} & (|\zeta| > |\cos(K/2)|) \\
-i \frac{1}{8t} \frac{1}{\sqrt{\cos^2 \frac{K}{2} - \zeta^2}} & (|\zeta| < |\cos(K/2)|).
\end{cases} \quad (5.6) \]

The important quantity in equations (4.20) and (4.21) is \( \arg R \). From (5.4) we see that if \( \zeta < -|\cos(K/2)| \), \( R \) is real and negative, while for \( \zeta > |\cos(K/2)| \) it is real and positive. For \(-|\cos(K/2)| \leq \zeta \leq |\cos(K/2)|\), \( R \) is pure imaginary with a negative imaginary part. So we find that

\[ \Phi(\zeta, K) = \arg R(\zeta, K) = \begin{cases} 
-\pi & (\zeta < -|\cos(K/2)|) \\
-\pi/2 & (|\zeta| \leq |\cos(K/2)|) \\
0 & (\zeta > |\cos(K/2)|).
\end{cases} \quad (5.7) \]
These values of $\Phi$ are plotted in figure 2 in the $\zeta, K/2$ plane. Integrating in the $K$-direction, we find for $F(\zeta)$

$$F(\zeta) = \frac{1}{N} \sum_K \arg R(\zeta, K) = \frac{1}{2\pi} \int_{0}^{2\pi} dK \Phi(\zeta, K)$$

$$= \begin{cases} -\pi & (\zeta < -1) \\ -\arccos \zeta & (-1 \leq \zeta \leq 1) \\ 0 & (\zeta > 1). \end{cases} \quad (5.8)$$

Using $\zeta$ instead of $E$ in (4.17) as integration variable we have

$$b_{\text{int}}^2 = \frac{1}{N} \int_{-1}^{1} d\zeta \left[ \rho_2(\zeta) - \rho_0^0(\zeta) \right] e^{-4\beta t (\zeta+1)}. \quad (5.9)$$

Due to the differentiation with respect to $z$ in (4,20) only values of $\zeta$ between $-1$ and $1$ contribute to the integral. From (5.8) we see that for $|\zeta| \leq 1$

$$\rho_2(\zeta) - \rho_0^0(\zeta) = -\frac{N}{\pi} \frac{\partial}{\partial \zeta} F(\zeta) = -\frac{N}{\pi} \frac{1}{\sqrt{1 - \zeta^2}} \quad (5.10)$$

Substituting this into (5.9) we find

$$b_{\text{int}}^2 = -e^{-4\beta t} I_0(4\beta t). \quad (5.11)$$

We now compare this with the result for an ideal lattice gas, given by (3.19)

$$b_0^0 = \frac{1}{2} b_1(2\beta) = \frac{1}{2} e^{-4\beta t} I_0(4\beta t) = -\frac{1}{2} b_{\text{int}}^2 \quad (5.12)$$

So the hard-core interaction giving rise to $b_{\text{int}}^2$ changes the value of $b_2$ from $b_0^0$ into its opposite, or in other words, the Bose value is turned into the Fermi value. This is exactly what has to be expected from the well known fact that a hard-core Bose gas in one dimension is equivalent to an ideal Fermi gas (for all virial coefficients).

The picture shown in figure 2 for the phase $\Phi$ has some general validity, in the sense that for sufficiently negative $\zeta$ one has $\Phi = -\pi$ while for $\zeta$ sufficiently positive $\Phi = 0$. In the zone in between one has for $d > 1$ in general a continuous transition from $-\pi$ to $0$, with eventually also zones with $\Phi = -\pi/2$.

**VI. THE TWO- AND THREE-DIMENSIONAL LATTICES**

The calculation of $b_2$ for hypercubic lattices in higher dimensions runs along the same lines as what was done in section 5 for one dimension. The band structure is now given by (again $a = 1$)

$$\varepsilon(\vec{k}) = \begin{cases} 2t(2 - \cos k_x - \cos k_y) & (d = 2) \\ 2t(3 - \cos k_x - \cos k_y - \cos k_z) & (d = 3). \end{cases} \quad (6.1)$$
In $d$ dimensions, $b_1(\beta)$ is simply given by

$$b_1(\beta) = \frac{1}{(2\pi)^d} \left[ \int_{-\pi}^{\pi} dk \, e^{-2\beta t(1-\cos k)} \right]^d$$

$$= e^{-2d\beta t [I_0(2\beta t)]^d}.$$  

(6.2)

As before, we rearrange the Brillouin zone such that its boundaries in $\vec{K}, \vec{k}$ space are convenient. The different components of $\vec{K}, \vec{k}$ are independent, and we have $0 \leq K_i \leq 2\pi$, $0 \leq k_x \leq \pi$, $-\pi \leq k_y, k_z \leq \pi$. The $k_x$-interval is halved to avoid counting the same (symmetric) state twice. The two-vacancy energy bands are given by

$$E_0(\vec{K}, \vec{k}) = 4t(2-\cos \frac{K_x}{2} \cos k_x - \cos \frac{K_y}{2} \cos k_y) \quad (d = 2),$$

$$E_0(\vec{K}, \vec{k}) = 4t(3-\cos \frac{K_x}{2} \cos k_x - \cos \frac{K_y}{2} \cos k_y - \cos \frac{K_z}{2} \cos k_z) \quad (d = 3).$$

(6.3)

We scale the parameter $z$ as

$$z = \begin{cases} 
8t(\zeta + 1 + i\epsilon) & (d = 2) \\
12t(\zeta + 1 + i\epsilon) & (d = 3),
\end{cases}$$

(6.4)

so that in both cases the energy band runs from $\zeta = -1$ to $\zeta = 1$.

In two dimensions we find with (5.3)

$$\mathcal{R}_2(\zeta, \vec{K}) = \frac{1}{(2\pi)^2} \int_{-\pi}^{\pi} dk_y \int_{-\pi}^{\pi} dk_x \frac{1}{8t} \left[ \zeta + i\epsilon + \frac{1}{2} \left( \cos \frac{K_x}{2} \cos k_x + \cos \frac{K_y}{2} \cos k_y \right) \right]^{-1}$$

$$= \frac{1}{32t\pi} \int_{-\pi}^{\pi} dk_y f(A, B) \sqrt{|A^2 - B^2|},$$

(6.5)

where

$$A = \zeta + \frac{1}{2} \cos \frac{K_y}{2} \cos k_y,$$

$$B = \frac{1}{2} \cos \frac{K_x}{2},$$

(6.6)

and

$$f(A, B) = \begin{cases} 
\text{sgn}(A) & (A^2 > B^2) \\
-i & (A^2 < B^2).
\end{cases}$$

(6.7)

The integral in (6.5) can be expressed in terms of complete elliptic integrals of the first kind (see appendix). This gives an analytic expression for $\mathcal{R}_2(\zeta, \vec{K})$.

This expression can also be used to find the result in three dimensions,

$$\mathcal{R}_3(z, \vec{K}) = \frac{1}{(2\pi)^3} \int_{-\pi}^{\pi} dk_z \int_{-\pi}^{\pi} dk_y \int_{0}^{\pi} dk_x \left[ z - 4t(3-\cos \frac{K_x}{2} \cos k_x - \cos \frac{K_y}{2} \cos k_y - \cos \frac{K_z}{2} \cos k_z) \right]^{-1}$$

(6.8)

or
\[ R_3(\zeta, \vec{K}) = \frac{1}{2\pi} \int_{-\pi}^{\pi} dk_x \int_{-\pi}^{\pi} dk_y \int_{0}^{\pi} dk_z \left[ \zeta + i\epsilon + \frac{1}{2}(\cos \frac{K_x}{2} \cos k_x + \cos \frac{K_y}{2} \cos k_y) \right]^{-1} \]
\[ = \frac{1}{2\pi} \int_{-\pi}^{\pi} dk_z R_2(\zeta, K_x, K_y), \]

where
\[ 8t(\zeta + 1) = 12t(\zeta + 1) - 4t(1 - \cos \frac{K_z}{2} \cos k_z). \]  

Using the analytic expression found for \( R_2 \), \( R_3 \) can be calculated by numerical integration of (6.9).

The next step is to obtain \( F(\zeta) \) as given in (4.21) by integrating over \( \vec{K} \). This is done numerically using Monte Carlo integration. The result is a function \( F(\zeta) \) that is equal to \(-\pi\) for \( \zeta < -1 \), where \( R_d \) is real and negative, equal to 0 for \( \zeta > 1 \), where \( R_d \) is real and positive, and that is in between these two values for \( \zeta \in [-1, 1] \), where \( R_d \) is complex. It follows from the symmetry of \( R_d \) that
\[ F(-\zeta) = -\pi - F(\zeta). \]  

Figures 3 and 4 show \( F(\zeta) \) for the square and cubic lattices.

Using \( F(\zeta) \), \( b_{\text{int}}^2 \) can be found from equations (4.17) and (4.20)
\[ b_{\text{int}}^2 = -\frac{1}{\pi} \int_{-1}^{1} d\zeta \left( \frac{\partial}{\partial \zeta} F(\zeta) \right) e^{-4\beta t(\zeta + 1)} \]  

Partially integrating, we find
\[ b_{\text{int}}^2 = -e^{-4\beta t} \left\{ e^{4\beta t} + \frac{4\beta t}{\pi} \int_{-1}^{1} d\zeta F(\zeta) e^{-4\beta t \zeta} \right\} \]
\[ = -e^{-4\beta t} \left\{ 1 - \frac{8\beta t}{\pi} \int_{0}^{1} d\zeta F(\zeta) \sinh 4\beta t \zeta \right\}. \]

To find \( b_{\text{int}}^2 \), the last equation can be numerically integrated for various values of \( \beta t \).

As before, we compare this with the result for the ideal lattice gas (3.13)
\[ b_2^0 = \frac{1}{2} b_1(2\beta) = \frac{1}{2} e^{-4\beta t} [I_0(4\beta t)]^d. \]

The ratio \( b_{\text{int}}^2 / b_2^0 \) is given by
\[ \frac{b_{\text{int}}^2}{b_2^0} = \frac{-2}{[I_0(4\beta t)]^d} \left\{ 1 - \frac{8\beta t}{\pi} \int_{0}^{1} d\zeta F(\zeta) \sinh 4\beta t \zeta \right\}. \]

For \( \beta t \to 0 \), this ratio goes to \(-2\). The second virial coefficient \( b_2 = b_2^0 + b_{\text{int}}^2 \) thus approaches \(-b_2^0 \) for high temperatures. This is indicative of fermionic behaviour, which is indeed what one would expect: at high temperatures, the only important contribution to the free energy of the system is the entropy involved in distributing a certain
number of hard core particles over the lattice. This is the same as for fermions. For $\beta t \to \infty$ the behaviour of $F(\zeta)$ depends on the behaviour of $F(\zeta)$ for $\zeta \to 1$. In two dimensions, $F(\zeta) \propto (1 - \zeta) / \ln(1 - \zeta)$, which leads to $b^2_{\text{int}} / b_0^2$ going to zero like $-1 / \ln(\beta t)$ as $\beta t \to \infty$. The same low-temperature behaviour is found for quantum hard disks [6]. In three dimensions, $F(\zeta) \propto (1 - \zeta)^2$, which gives $b^2_{\text{int}} / b_0^2 \propto -1 / \sqrt{\beta t}$ for $\beta t \to \infty$, just as for quantum hard spheres [6]. So in both cases, $b_2$ approaches the value for the ideal Bose gas at low temperatures. This shows that when the thermal wavelength (3.9) exceeds the lattice constant $a (= 1)$, the effects of the Bose statistics starts to dominate. Plots of $b_2 / b_0^2$ versus $\beta t$ for two and three dimensions are given in figures 5 and 6.

VII. CONCLUSION

The pressure and density of a gas of quantum particles on a lattice can be expressed in fugacity expansions,

$$p = \frac{1}{\beta v_0^0} \sum_{\ell=1}^{\infty} b_\ell e^{-\ell \beta \Delta}, \quad (7.1)$$

$$n = \frac{1}{v_0^0} \sum_{\ell=1}^{\infty} \ell b_\ell e^{-\ell \beta \Delta}. \quad (7.2)$$

For bosons with a hard-core interaction, the first two coefficients, $b_1$ and $b_2$, can be calculated. It turns out that the effect of the hard-core interaction on $b_2$ depends strongly on temperature and on the transfer integral $t$. For small $\beta t$ it is fermionic in character, as far as $b_2$ is concerned. For large $\beta t$ the effect of the hard-core disappears, and only the Bose character remains.

The system of vacancies that exists in solid $^4\text{He}$ should behave to a good approximation like this simple model: the vacancies move through the crystal lattice by a tunneling process, they are bosons, and they have a hard-core repulsion. The gas of vacancies has been experimentally studied by probing the attenuation and the velocity shift of sound in a $^4\text{He}$ crystal [5]. Very pure hcp Helium was used, so that the effects of both the phonons and the delocalized, bosonic vacancies could be observed. Treating the vacancies as a gas of free particles, it was seen that they obey Bose statistics. For the expressions (7.1) and (7.2) this means that not only the first term, but at least also the second is experimentally observable. One can use the expressions (3.14) and (3.15) for a gas of free, ideal bosons to estimate the order of magnitude of the various terms in (7.1) and (7.2). One finds that

$$b^0_\ell = v_0 / \ell^{3/2} A_3^3,$$

so that

$$p = \frac{1}{\beta \lambda^3} \sum_{\ell=1}^{\infty} \frac{e^{-\ell \beta \Delta}}{\ell^{3/2}}, \quad (7.3)$$

$$n = \frac{1}{\lambda^3} \sum_{\ell=1}^{\infty} \frac{e^{-\ell \beta \Delta}}{\ell^{3/2}}. \quad (7.4)$$
Using the value $\Delta/k_B = 0.71K$ found in table 1, and the maximum temperature $T = 0.85K$ at which the experiments were performed, this shows that the ratio between the second and first terms in (7.4) is 0.15, and that between the third and first terms is 0.04.

If experiments can be done that detect the contribution of 15% of the second term in (7.4), it is also possible to detect the effects of the hard-core on the coefficient $b_2$ in (7.2). As can be seen from figure 6, it varies considerably with $\beta t$, and it can even change sign. However, it is difficult to extract this information from the attenuation experiment, since there it is not clear what exactly the relation between the measured quantities and the vacancy density is. It would be necessary to directly measure, say, the pressure due to the vacancies, and then compare this with (7.4). In such an experiment it would be crucial to take the hard-core effects into account. Work is in progress to calculate the coefficient $b_2$ for the hcp lattice; however, the results are not expected to differ much from those given for the simpler cubic lattice here.

The results for the two-dimensional system might have relevance for low-density $^4$He films on textured substrates. There, the presence of adsorption sites localizes the Helium particles on the points of a two-dimensional lattice, defined by the substrate. Thus the tight-binding approach used in this paper becomes applicable. The adsorption sites cannot hold more than one particle, so that the hard-core repulsion is also present.

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The integral (6.3) can be expressed in terms of elliptic integrals as follows.[10,11] $R_2$ is equal to

$$R_2(\zeta, \vec{K}) = \frac{1}{16t\pi} \int_{A^2 > B^2} dk_y \frac{\text{sgn}(A)}{\sqrt{A^2 - B^2}}$$

$$- \frac{i}{16t\pi} \int_{A^2 < B^2} dk_y \frac{1}{\sqrt{B^2 - A^2}}, \quad (5)$$

where $k_y \in [0, \pi]$. On making the substitution $p = \cos k_y$, and writing

$$A^2 - B^2 = \cos^2(K_y/2) \frac{1}{4}(q_1 - p)(q_2 - p), \quad (6)$$

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where
\[ q_1 = -2\zeta + \cos(K_x/2), \quad q_2 = -2\zeta - \cos(K_x/2), \] (7)
we end up with integrals of the type
\[ \int dp \frac{1}{\sqrt{|w|}}, \] (8)
where \( w = (1-p)(1+p)(q_1-p)(q_2-p) \) and \( p \in [-1,1] \).

By defining
\[ q_- = \min(q_1, q_2), \quad q_+ = \max(q_1, q_2), \] (9)
we see that values of \( p \in [q_-, q_+] \) give \( A^2 < B^2 \), and thus contribute to the imaginary part of \( \mathcal{R}_2 \). Values of \( p \) outside this interval give \( A^2 > B^2 \) and thus contribute to the real part, for \( p < q_- \) with \( \text{sgn}(A) = -\text{sgn}(\cos(K_y/2)) \), and for \( p > q_+ \) with \( \text{sgn}(A) = \text{sgn}(\cos(K_y/2)) \). The result is a sum of integrals of the type (8) between limits that are consecutive zeroes of \( w \). These integrals can all be expressed in terms of the complete elliptic integral of the first kind, \( K(x) \) [12],
\[ K(x) = \int_0^1 \frac{1}{\sqrt{(1-p^2)(1-x^2p^2)}} \quad (x \in [0,1]). \] (10)

There are several cases to be considered:

1. \( q_-, q_+ < -1 \) (1a) or \( q_- < -1, q_+ > 1 \) (1b)
\[ \mathcal{R}_2(\zeta, \vec{K}) = \frac{\epsilon}{4t\pi \cos(K_y/2)} \frac{K(r_1)}{(q_+ - 1)(q_- + 1)}, \] (11)
where \( \epsilon = + \) for (1a) and \( \epsilon = - \) for (1b).

2. \( q_- < -1, q_+ > 1 \) or \( q_- < -1, q_+ \in [-1,1] \)
\[ \mathcal{R}_2(\zeta, \vec{K}) = \frac{-i}{4t\pi \cos(K_y/2)} \frac{K(r_2)}{\sqrt{(q_+ + 1)(1 - q_-)}}, \] (12)

3. \( q_- < -1, q_+ \in [-1,1] \) (3a)
or \( q_- \in [-1,1], q_+ > 1 \) (3b)
\[ \mathcal{R}_2(\zeta, \vec{K}) = \frac{\epsilon}{4t\pi \cos(K_y/2)} \frac{K(1/r_1)}{\sqrt{2(q_+ - q_-)}} - \frac{i}{4t\pi \cos(K_y/2)} \frac{K(1/r_2)}{\sqrt{2(q_+ - q_-)}}, \] (13)
where \( \epsilon = + \) for (3a) and \( \epsilon = - \) for (3b). In the above, \( r_1 \) and \( r_2 \) are given by
\[ r_1 = \sqrt{\frac{2(q_+ - q_-)}{(q_+ - 1)(q_- + 1)}} \quad \quad r_2 = \sqrt{\frac{2(q_+ - q_-)}{(q_+ + 1)(1 - q_-)}} \]

\[(14)\]

[1] A.F. Andreev, in: *Progress in Low Temperature Physics*, Vol. 8 (D.F. Brewer, ed.), North Holland, Amsterdam, 1982.
[2] A.F. Andreev and I.M. Lifshitz, Zh. Exp. Teor. Fiz. 56 2057 (1969) [Sov. Phys. JETP 29 1107 (1969)].
[3] T. Matsubara and H. Matsuda, Prog. Theor. Phys. 16 569 (1956); H. Matsuda and T. Matsubara, Prog. Theor. Phys. 17 19 (1957).
[4] G.A. Lengua and J.M. Goodkind, J. Low Temp. Phys. 79 251 (1990).
[5] K. Huang, *Statistical Mechanics* (2nd edition), Wiley, New York, 1987.
[6] R.L. Siddon and M. Schick, Phys. Rev. A 9 907 (1974); W.G. Gibson, Mol. Phys. 49 103 (1983).
[7] G.E. Uhlenbeck and E. Beth, Physica 3 729 (1936).
[8] S. Steed, to be published.
[9] J.J. Rehr and M. Tejwani, Phys. Rev. B 20 345 (1979).
[10] T. Morita and T. Horiguchi, J. Math. Phys. 12 986 (1971).
[11] E.W. Montroll, in: *Proceedings of the third Berkeley Symposium on Mathematical Statistics and Probability*, Vol. 3 (J. Neyman, ed.), University of California Press, Berkeley, 1955.
[12] I.S. Gradshteyn and I.M. Ryzhik, *Table of Integrals, Series, and Products*, Academic Press, San Diego, 1980.

FIG. 1. The Brillouin zone for the one-dimensional problem. Domains I and III' are the Brillouin zone used in the calculation.

FIG. 2. The phase $\Phi(\zeta, K)$ in the $\zeta, K/2$ plane.

FIG. 3. The function $F(\zeta)$ for the square lattice. The only values of interest are those between $\zeta = -1$ and $\zeta = 1$, where $F(\zeta)$ is not constant and varies from $-\pi$ to 0. Only positive values of $\zeta$ are shown, since $F(\zeta)$ for negative $\zeta$ can be found using the antisymmetry of that function around $\zeta = 0$ and $F(\zeta) = -\pi/2$.

FIG. 4. The function $F(\zeta)$ for the cubic lattice.
FIG. 5. The ratio $b_2/b_2^0$ as a function of $\beta t$ for the square lattice. For high temperatures (small $\beta t$), $b_2 = -b_2^0$, which is the same value as for fermions. For lower temperatures its value crosses over to $b_2 = b_2^0$, the ideal boson value.

FIG. 6. The ratio $b_2/b_2^0$ as a function of $\beta t$ for the cubic lattice. Note that the cross-over to the ideal boson value is much faster than for the square lattice.
\[ F(\zeta) \]

\[ 0 \quad \frac{-\pi}{2} \]

\[ 0 \quad 0.2 \quad 0.4 \quad 0.6 \quad 0.8 \quad 1 \]

\[ \zeta \]
$F(\zeta)$
