Path Integral Calculations of Vacancies in Solid Helium

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Abstract

We study properties of vacancies in solid ^{4}He using Path Integral Monte Carlo. We find, in agreement with other calculations, that the energy to create a single vacancy is 11.5K and is monotonic with the number of vacancies. If more the a few percent of the system becomes vacant, we find the system becomes unstable to melting. We show the number of exchanges in the system is increased by vacancies and how the underlying lattice is altered by the presence of a vacancy. We also examine the efficacy of using a tight binding hamiltonian to describe the vacancy in the crystal, show that vacancies are attractive, and find values for the effective mass and inter-vacancy attraction.

Key words:
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1. Introduction

Because of the quantum nature of ^{4}He, it exhibits a number of anomalous properties including superflow and Bose-Einstein condensation (BEC) in the liquid phase, the ability to stay liquid at zero temperature, and a large zero point motion in the solid. Recent work by Kim and Chan[5] have also found indications of non-classical rotational inertia (NCRI) in solid ^{4}He. The physics behind this behavior is still unclear.

One initially plausible explanation for this effect was that the wave function that represented the ground state of ^{4}He exhibited superfluidity or Bose-Einstein Condensation (BEC). Recent theoretical calculations indicate that the bulk, commensurate, equilibrium solid (either in the ground state or at the finite temperature of the experiments) has neither ODLRO [7,8] nor superfluidity [6]. Many of these calculations were done at a fixed particle number commensurate with a lattice that has no vacancies in accordance with the original suggestion of Andreev and Lifshitz[9] and Chester [11]. This leaves open the possibility that the ground state of Helium has zero point vacancies [12] and that these vacancies induce supersolidity. Another possibility is that although a single vacancy may be energetically unstable, a gas of vacancies may have lower energy than the commensurate crystal [13].

Even if vacancies do not play an important role in the equilibrium state of solid ^{4}He at sufficiently low temperatures, it is plausible they may be prevalent in experiments at higher temperatures. In fact, recent experimental results have indicated that annealing of the crystal results in an elimination or reduction of the supersolid signal [14]. It is a reasonable conjecture that the pre-annealed crystals have a number of defects including vacancies, dislocations and grain boundaries, while the post-annealed crystal has fewer of these "non-equilibrium" defects.

Hence, the understanding of vacancies in solid helium is of great interest. Beyond these considerations, there has been little work calculating the role that quantum vacancies play in realistic materials. A classical model of vacancies is insufficient because the quantum nature of the system allows for delocal-

ization and Bose condensation of vacancies. In this work, we will explore properties of vacancies using Path Integral Monte Carlo (PIMC). In Section 2 we will discuss our method. In Section 3 we will discuss the energy costs of introducing vacancies into the system. In Section 4 we will examine the way in which an introduction of a vacancy distorts the underlying solid lattice. In Section 6 we discuss the relation of the vacancy system to tight binding hamiltonians. Finally in Sections 7 and 8 we calculate the effective mass of the vacancy and the attraction between vacancies, respectively.

2. Path Integral Monte Carlo

Path Integral Monte Carlo (PIMC) is a numerical technique that exactly calculates properties of bosonic equilibrium systems by integrating over the density matrix. The PIMC calculation's input are the interatomic Helium potential V , taken to be the Aziz 1995 semi-empirical form[10], the number of atoms, the size of the periodic box, and the temperature. We integrate over the density matrix

$$\rho(R) = \langle R| \exp(-\beta H)|R\rangle$$

where

$$H = -\lambda\nabla^2 + V(R).$$

Calculations are done at temperatures from $0.25K$ to $1K$ in a periodic box with a density of 0.02862 ptcl/$\mathring{A}^3$. (This corresponds to a pressure of approximately 26.7 bars, just above the melting density where quantum effects should be maximized in the crystal.) The aspect ratio of the simulation box is chosen so that an hcp lattice of 180 lattice sites is commensurate with the periodic boundary conditions. The number of particles in the system is then set by the number of vacancies desired in the system. The only effect of these lattice sites is to seed the initial Monte Carlo configurations and to define the Wigner-Seitz cells of the system. Calculations reported here were done with PIMC++, a C++ code that implements the algorithms described in Ceperley[3].

3. Energy of Adding Vacancies

We start by calculating the energy cost of introducing a single vacancy into an equilibrium crystal. Anderson et al.[12] argue that the equilibrium state of solid ^{4}He could be incommensurate. If the system is incommensurate, then we expect that the crystal with a vacancy will have a lower free energy than one without. At low enough temperature, and for a low density of vacancies, the concentration of vacancies is given by $\exp(-\beta\Delta E)$ where ΔE is the energy cost of a vacancy. We calculate the energy cost for introducing a vacancy into the system at T=0.5K. We make the distinction between the energy cost of introducing a vacancy into a quantum "boltzmannon" crystal (where there are no permutations that implement Bose statistics) and introducing a vacancy into a bosonic quantum crystal. While at zero temperature, statistics should not matter, at finite temperature the internal energy will depend on statistics.

We find the energy cost of introducing a vacancy into a bosonic crystal is $11.5\pm1.1K$, which is comparable with Pollet [1] who gets a value of $13.0 \pm 0.5K$ for the vacancy energy and to a variational estimate of Pederiva et.al [2] of $11.6\pm2.0K$ for a slightly different density. The delocalization caused by turning on the bose statistics results in a statistically insignificant change in the perfect crystal but approximately a 2-3 Kelvin drop in the system with a vacancy. This is likely a result of the fact that although permutations are minimal in the perfect crystal, the system with a vacancy allows for many permutations allowing for a drop in the energy.

Dai et al. [13] propose that the energy cost for introducing a single vacancy might be energetically costly, but that the energy might drop for a non-zero concentration of vacancies. To test this, we examine the energy of the system as a function of vacancy concentration. This is shown in figure 1. The energy cost for introducing vacancies is positive and monotonic in the number of vacancies up until the point where the system begins to destabilize and melt. This is evidence that the normal equilibrium crystal is commensurate and there is no indication that a non-zero finite concentration of vacancies is energetically favorable in the ground state of ^{4}He. During the PIMC calculation, if more than 2% (4 out of 180 sites) were vacant, the system collapsed into a liquid-like phase. Such unstable systems are marked with arrows on figure 1. The stability of the crystal was determined by monitoring the structure factor for k-vectors close to the hcp reciprocal lattice vector. The structure factor is shown in fig. 2 for calculations of 4 and 5 vacancies. It is seen that the addition of one vacancy causes the maximum value of $S(k)$ to drop from roughly 40 to less than 10.

This instability may be relevant to a form of crystal destabilization seen in the experiment on solid

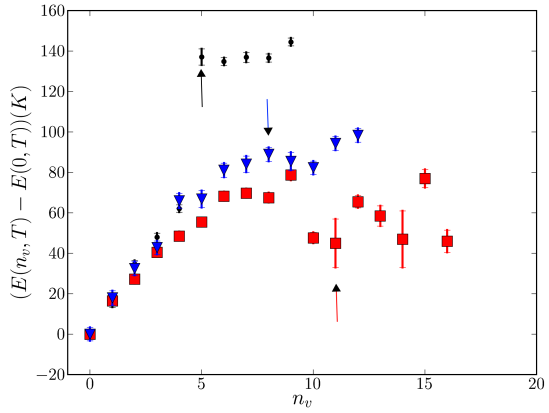

Fig. 1. Energy difference as a function of vacancy number n_v in a system of $n_s = 180$ lattice sites for 2K (black circles), 1K (red squares) and 0.5K (blue triangles). $E(n_v, T)$ is the internal energy, with the density held fixed. The arrows indicate the onset of melting as signalled by the loss of Bragg peaks.

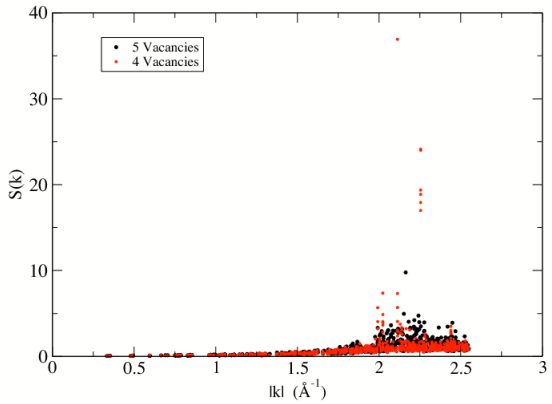

Fig. 2. Structure factor for a system at $2K$ with either 4 (red stars) or 5 (black circles) vacancies in the system. Points at the same value of k are in different directions.

^{4}He by Toennies et al.[4] In their experimental system they "inject vacancies" through a hole in solid ^{4}He. As these vacancies are injected into the system, there are macroscopic jumps in the pressure of the system which indicates a restructuring or melting/refreezing of the crystal, consistent with what is seen in our simulation.

4. Lattice Distortion due to a Vacancy

If a hole is introduced into the lattice and the lattice does not relax, there is a unique lattice site whose Wigner-Seitz cell is devoid of atoms. However, with zero point motion, the lattice will relax and the missing density will be distributed in neighboring lattice sites. The size of this distortion is a property of the quantum vacancy. If we average long enough, since our system has translation invariance, the density will be uniformly spread through the simulation cell. In order to say something about the size of the vacancy at finite temperature, we proceed as follows. Let $\rho_k^N = \sum_i \exp(ik \cdot r_i)$ be the Fourier transform of the instantaneous density for a system with N atoms. Then

$$\rho(\boldsymbol{r}) = \frac{1}{(2\pi)^3} \int d^3\boldsymbol{k} \exp(-i\boldsymbol{k} \cdot \boldsymbol{r}) \rho_{\boldsymbol{k}}.$$

The structure factor is

$$S(k) = \frac{1}{N} \langle |\rho_k|^2 \rangle.$$

An estimate of the density of a vacancy is obtained by subtracting the density of a perfect crystal from that of a single vacancy. Let us assume that the k-space density of the vacancy is $\delta\rho \equiv \rho^N - \rho^{N-1}$. Then

$$|\rho_k^{N-1}|^2 - |\rho_k^N|^2 = |\delta\rho_k|^2 - 2Re\rho_k^N \delta\rho_k.$$

We note that $Re[\rho_k^N \delta\rho_k]$ can be neglected since ρ_k^N is small for k not a reciprocal lattice vector and has a random phase. Therefore, we have

$$\delta\rho_k \approx [(N-1)S_k^{N-1} - NS_k^N]^{1/2}$$

where we have assumed that the vacancy is centered at the origin and is hence real. Figure (3) shows the value of $\delta\rho(r)$ for a quantum system of ^{4}He. Note the vacancy has a "negative presence" from 1.6 to 3.2 $\mathring{A}$; *i. e.* an increase in density surrounding the vacancy caused by other particles expanding and encroaching upon the vacant area. If we define the vacancy by the missing density (i.e. ignoring the extra density that is added onto the system), the primary site of the vacancy has 9% of the total density of the vacancy, the 12 nearest neighbors contain 32% of the density and the second nearest neighbors contain the rest of the density.

One can also recognize the distortion of the lattice by examining the particle locations with respect to their corresponding lattice sites. Because ^{4}He has a large zero point motion, though, looking

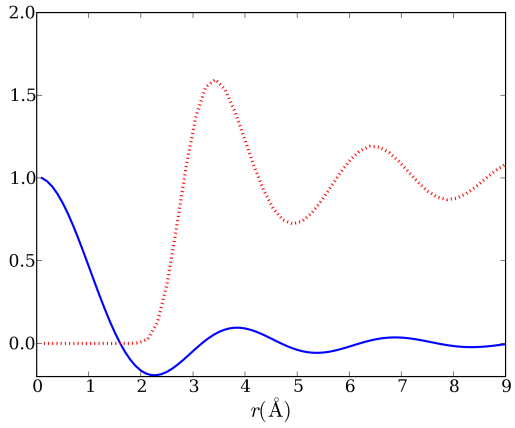

Fig. 3. The solid line (blue) represents the value of $\delta\rho(r)$ for the vacancy. Values below 0 represent an increase in density or a "negative" vacancy. The dotted line (red) is the pair correlation function of a perfect helium crystal.

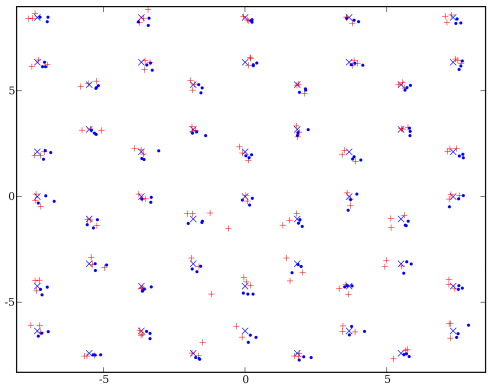

Fig. 4. Shown is a snapshot of a 2d projection from the 3d hcp crystal. The x's (blue) are the location of the lattice sites. The dots (blue) are the centroids of the paths of a perfect crystal. The +'s (red) are the centroids of the paths of a crystal with a single vacancy. Many particles in the system with a vacancy have migrated significantly off their respective lattice sites.

at a snapshot of any imaginary time slice on the Helium "path" is very noisy. Instead, one can look at a snapshot of the centroids of the paths (i.e. the center of mass of each polymer) as shown in Figure 4; compared with the commensurate crystal, in the situation with a single vacancy the surrounding atoms drift significantly away from their respective lattice sites

The presence of a vacancy also significantly affects the cyclic permutations that are generated by the method. In the path integral method, paths permuting with each other are how Bose statistics are implemented and their presence reflects exchange. Figure 5 shows the number of permutations of a given size. When the system is commensurate, there are only a few 2,3, and 4 particle permutations. On the other hand, when a vacancy is introduced into the system, permutations up to size 14 exist and the number of exchanges increases drastically.

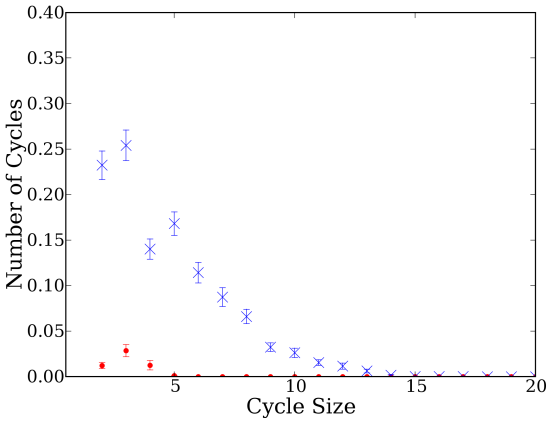

Fig. 5. Number of paths permuting onto each other to form a cycle of a given size. The dots (red) are for the perfect crystal and the x's (blue) are for the system with a single vacancy.

5. Vacancy-Interstitial Definition

To further explore properties of the vacancy, we need to specify where the vacancy is located by assigning it to a lattice site. A variety of such definitions exist. Galli and Reatto [2] define a vacancy as a Wigner-Sietz cell that is empty and has no doubly occupied nearest neighbors. In ref. [15] vacancies are located by removing pairs of close particles and lattice sites from the system in a "greedy" fashion; the last remaining lattice site is designated as the location of the vacancy. Alternatively, one may define a vacancy in the following way. First, one matches each particle to at most one lattice site such that the sum of the distance squared between particles and lattice sites is minimized. Suppose that there are M lattice sites and $N < M$ helium atoms and let r_i be the instantaneous position of atom i. Define

$$\chi_P = \sum_{i=1,M} d_{i,P_i} \qquad (1)$$

Here P is a permutation of the integers $\{1, \ldots, M\}$, $d_{i,j}$ is the squared distance between atom i and lattice site j and define $d_{i,j} = 0$ for $i > N$. We then determine P such that χ_P is minimized. [1] We choose to use this last definition because it finds the global minimum of χ, not necessarily a local one. It allows us to produce a path for the vacancy as a function of imaginary time. Where fluctuations might allow a Monte Carlo configuration to have varying number of vacancies, this definition always uniquely defines the number of vacancies equal to the number of sites minus the number of atoms. The presence of a nearby double occupation, does not exclude the designation of a site as being vacant.

6. Mapping onto a tight-binding model

The simplest model for the energetics of a dilute gas of vacancies is a tight binding lattice model whose Hamiltonian is:

$$H = t \sum_{\langle i,j \rangle} c_i c_j^\dagger + V_{in} \sum_{i,j \in plane} c_i c_i^\dagger c_j c_j^\dagger + V_{out} \sum_{i,j \notin plane} c_i c_i^\dagger c_j c_j^\dagger \quad (2)$$

having a nearest neighbor hopping and a nearest neighbor interaction term. In this model, two vacancies cannot occupy the same site. Here c_i creates a vacancy at lattice site i and $t_{i,j}$ is the hopping matrix element between nearest neighbor sites (i, j). In an hcp lattice the hopping can be different in and out of the basal plane: t_{in} and t_{out}. Here V_{in} and V_{out} are nearest neighbor interactions.

However, there are limitations to this model. Let us now contrast the picture of what is going on in the lattice system with what is going on in the continuum system. In the lattice system, when the vacancy "hops" in imaginary time from one lattice site to another lattice site, it "loses" memory of where it has come from. In the continuum case, though, the vacancy may "hop" from one lattice site to another lattice site by moving a fraction of the inter-particle distance so that the missing density now mainly inhabits a neighboring Wigner-Seitz cell. After this hopping there is a resident memory of where it has come from and so it is significantly less costly to hop back to its former lattice site than to hop to another nearest neighbor. In figure (6) we plot the log of the probability the vacancy has moved from 0 to r in imaginary time $2\tau = 0.05$. We show model systems with effective masses ranging from $0.1 m_{\mathrm{He}}$ to $10 m_{\mathrm{He}}$ and see that they all have a different qualitative behavior from the actual vacancy in the PIMC data. Although we can't rule out that this is an artifact of how the vacancy is defined, we believe that this gives strong evidence that the wide band gap hamiltonian is a poor representation of a vacancy in solid Helium.

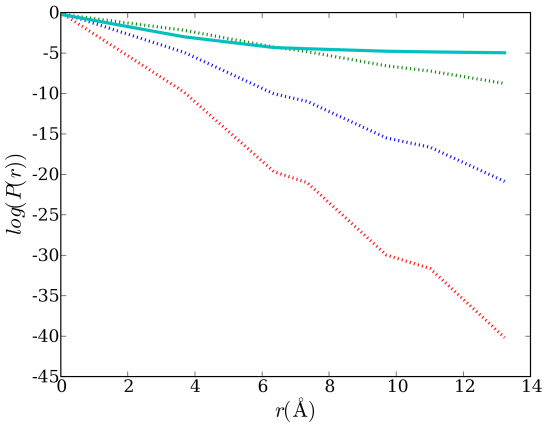

Fig. 6. Log of the probability the vacancy is on a lattice site r in the basal plane after time $2\tau = 0.05$. Dashed lines represent model systems with effective masses of 0.1 (top dashed line), 1(middle dashed line) and 10 (bottom dashed line) times the mass of Helium. Solid line is PIMC data. Note that it is qualitatively inconsistent with the model.

7. The Effective Mass of a Vacancy

One aspect unique to a quantum crystal is the ability for it to delocalize. In path integrals, the signature of delocalization of the vacancy is the vacancy's presence on different lattice sites at different imaginary times. The hopping rate is given by t and is inversely proportional to the effective mass of the vacancy. In this section, we show that the vacancy has an effective mass of approximately 0.15 times that of a Helium atom. Naively, an effective mass smaller than a helium atom seems unusual because it would seem that moving to a neighboring lattice site would require pushing the Helium atom that is

[1] This problem is the linear sum assignment problem and is solved exactly in order M^3 operations with the Hungarian method [18]. The vacancies are then located at lattice sites $Z_{P_{N+1}} \ldots Z_{P_M}$. Interstitials (i.e. $M < N$) can be defined by interchanging the role of atoms and lattice sites in the above description. The definition $d_{i,j}$ as the squared distance rather than some other metric means that the hypervolumes are bounded by hyperplanes, not by curved surfaces.

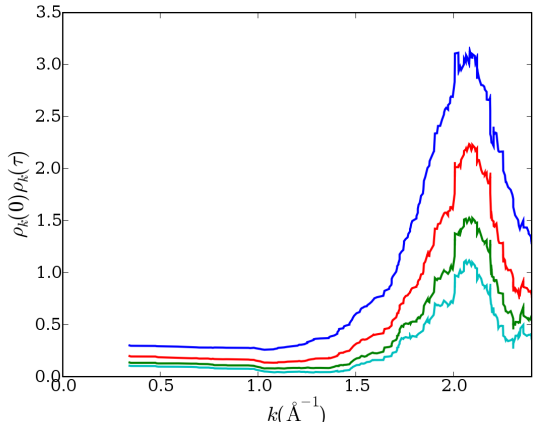

Fig. 7. $\rho_k(0)\rho_k(\tau)$ for $\tau = 0.1, 0.2, 0.3, 0.4$ (respectively descending). Curves have been smoothed through the use of a sliding window.

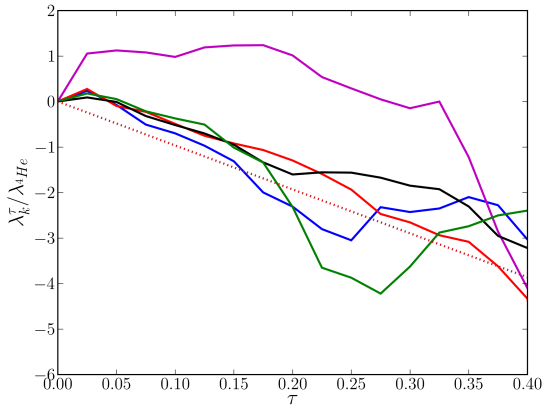

Fig. 8. Sample of 5 large k-values (jagged lines) as a function of τ. The slopes of these lines should be λ_k. The dotted line is the best fit to these slopes.

currently in that site. This reasoning is misleading, though, and a result of ignoring that the system is actually on a continuum as opposed to a lattice. In the continuum, a Helium atom need move only fraction of a lattice site for the vacancy to swap onto another lattice site. Below we will use two different techniques to calculate the effective mass of the vacancy.

7.1. *Measuring via $F_k(\tau)$*

One method for establishing the effective mass for a vacancy is through the imaginary time dynamic structure factor $F_k(\tau)$ Note that

$$\delta F_k(\tau) = F_k^N(\tau) - F_k^{N-1}(\tau)$$

where $F_k(\tau) = \rho_k(0)\rho_{-k}(\tau)$. Figure 7 shows $\delta F_k(\tau)$ for $\tau \in \{0.1, 0.2, 0.3, 0.4\}$. Defining λ_k^τ by $\delta F_k(\tau) = \delta S_k \exp(-\lambda_k^\tau k^2 \tau)$ we can calculate the effective mass of the vacancy. For each k, we fit the values of $\lambda_k^\tau/\lambda_{^4\text{He}}$ versus τ to a line for a range of $\tau \in (0.025, 0.25)$, a sampling of which is seen in figure 8. Figure 9 plots the slope of these lines (calculated by doing a chi-squared fit) versus k^2. By averaging the value of this mass at large k, we find an effective mass that is 0.15 times that of a helium atom.

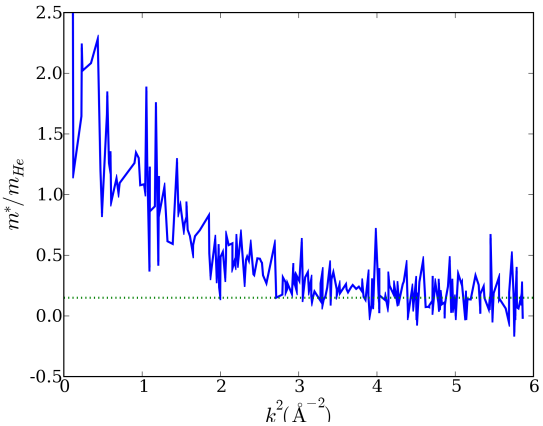

Fig. 9. Values of $m^*/m_{^4He}$ as a function of k^2. The y-axis is in units of the ^4He mass. The dotted line is a guide to the eye indicating the average of the large k values.

7.2. *Effective mass from the tight binding model*

In the previous subsection, we calculated the effective mass of the vacancy by looking at the dynamic structure factor. In this subsection, we will estimate the effective mass of the vacancy using the imaginary time position-position correlation functions in both the continuum (with the exact Hamiltonian) and the lattice model (with an effective Hamiltonian as described in Eq. (2)). Although there is reason to believe that this lattice model can not accurately represent the continuum system, it should, nonetheless, be an effective approximation at small τ and small r. To get the imaginary time correlation functions for the model system, we diagonalize the model hamiltonian and calculate the correlation functions as a function of the eigenfunctions and eigenvalues of the system for a given value of the inverse temperature β. We use the correlation functions $D(r) = \langle (r_{vac}(0) - r_{vac}(\tau))\delta(r - (r_{vac}(0) - r_{vac}(\tau))\rangle$ where

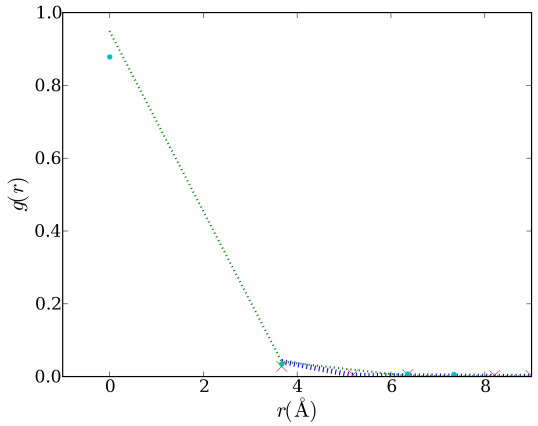

Fig. 10. Comparison of the PIMC data (blue dots (basal plane) and x's (AB sites)) with the model (dotted lines) for a hopping parameter of $t = 1.6K$ as found in in the analysis of $F_k(\tau)$

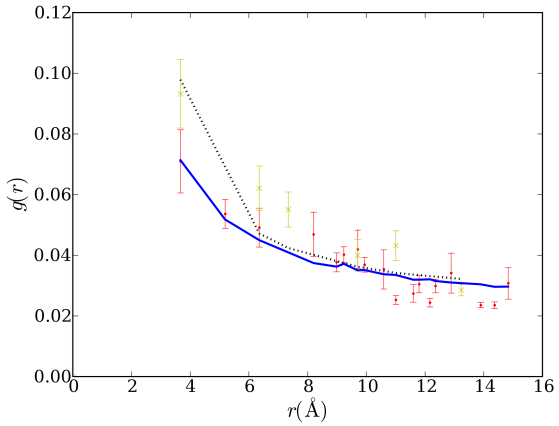

Fig. 11. Pair correlation functions of 2 vacancies at 1 K. The dots (red) are for AB sites. The x's (yellow) are for the in-plane sites. The blue (black) lines are the best parameter model fits for the in-plane (AB) sites.

$r_{vac}(\tau)$ is the position of the vacancy at imaginary time slice τ. We separately calculate correlation functions for sites in the basal plane and on the AB lattice. Once we determine the hopping parameter by fitting the correlation function, we can map onto a mass. To do this, we rewrite the Laplacian as a 12 point formula on the hcp lattice and find that $\lambda\nabla^2 = \lambda/(2a^2) = t$ giving us that $m^* = 1/(4a^2t)$ where a is the nearest neighbor distance. We fit the hopping parameter t in two ways. To begin with, we fit the entire correlation function for $\tau = 0.025$ by minimizing the chi-squared difference of the correlation function of our model lattice system and the continuum path integral results. This gives an effective mass of approximately 0.08 m_{He_4}. We should note that although this is the best fit, it does not fit the data particularly well. Because there is reason to believe that the data at large r is an especially bad fit to the model (since at small τ it shouldn't hop there), we also fit only the ratio between nearest neighbor basal plane hopping (this cancels out normalization effects influencing our fit). Doing this gives us an effective mass of 0.11 m_{He_4}. These values are in reasonable agreement with the calculative using $F_k(\tau)$. See figure 10 for a comparison of our our model with our continuum answer with a t of 1.6K.

8. Vacancy Attraction

Beyond the effective mass, another critical ingredient to understanding the behavior of vacancies in solid Helium is the vacancy-vacancy interaction. A strong attractive interaction will cause clusters of vacancies (or voids) to form. Unlike voids in metals, which might contain a gas, these voids could collapse. The void formation would stymie the role of vacancies in promoting superflow as there would be domains of perfect bulk crystal which have been shown to be a normal solid[6]. On the other hand, a repulsive or very weak attractive interaction might allow a gas of delocalized vacancies to permeate the solid Helium background. Although previous calculations [15] have looked at attraction of multiple vacancies, to date only variational calculations [16] have looked at 2 vacancies and no one has calculated the vacancy-vacancy interaction term. This is particularly important because it allows us to calculate the relevant interaction terms in our tight binding hamiltonian. As can be seen from the vacancy-vacancy correlation function in figure 11, our calculations clearly show the vacancies are attractive.

Although there is a noticeable attraction, the pair correlation functions does not seem to indicate the divacancy is bound at 1K since the probability of being arbitrarily far away is still non-zero. To quantify this attraction, we fit the parameters to our tight binding model. For our model of 2 vacancies, we make a change of basis to the center of mass coordinates on the lattice. We then calculate $g\left(|\boldsymbol{r}|\right)$for the basal plane and AB sites in the lattice model and path integral calculation and choose parameters V_{in}, V_{out} that minimize the squared difference between these results). Fitting these two parameters we get $V_{\mathrm{in}}/t = 7.3 \pm 0.5$ and $V_{\mathrm{out}}/t = 4.5 \pm 0.5$. Given the

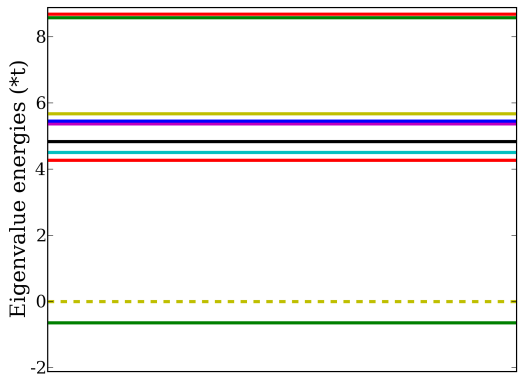

Fig. 12. Eigenvalue energies (as a multiple of the hopping parameter t). All values below 0 are bound. We note that there is a single bound state in the system.

value of these lattice parameters, we are then able to calculate the energy spectrum of the system as shown in figure 12. There appears to be only a single (s-state) bound eigenvalue. The binding energy of a di-vacancy is approximately $0.47t$. Using our value for the hopping parameter $t = 1.6K$ we find the binding energy is 0.75 K. Hence, we expect at low temperatures, any free vacancies, would form bound di-vacancy states.

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