The Abdus Salam International Centre for Theoretical Physics

United Nations Educational, Scientific and Cultural Organization

IAEA International Atomic Energy Agency

1929-3

Advanced School on Quantum Monte Carlo Methods in Physics and Chemistry

21 January - 1 February, 2008

PIMC 2 Bosons.

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Bosonic Path Integrals

1. Overview of effect of bose statistics
2. Permutation sampling considerations
3. Calculation of superfluid density and momentum distribution.
4. Applications of PIMC to liquid helium and helium droplets.
5. Momentum distribution calculations
6. MAXENT calculation of real time information from imaginary time correlation functions.

Quantum statistics

- For quantum many-body problems, not all states are allowed: allowed are totally symmetric or antisymmetric. Statistics are the origin of BEC, superfluidity, lambda transition.
- Use permutation operator to project out the correct states:

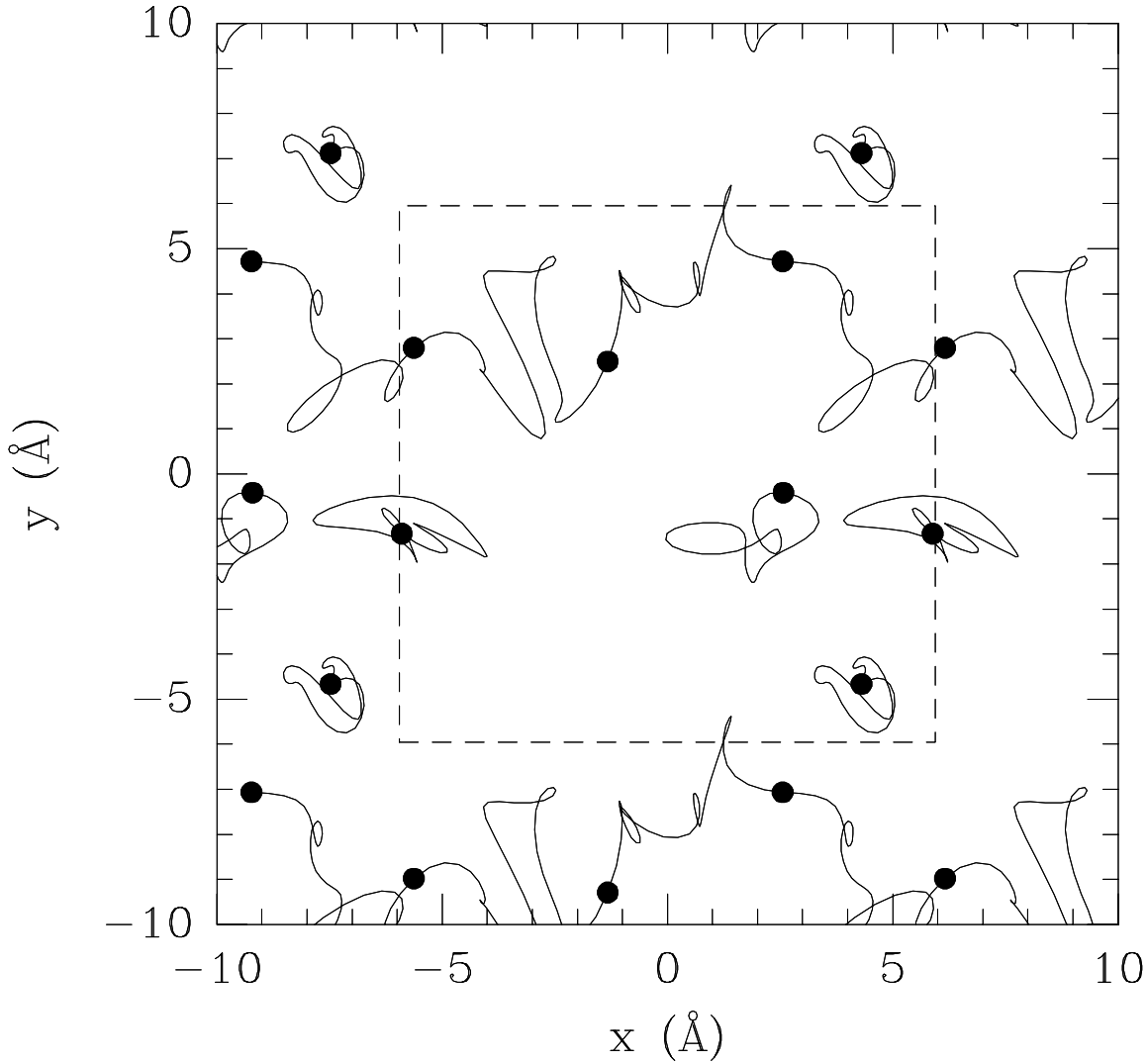
$$\hat{P} f(R) = \sum_{p=1}^{N!} \frac{1}{N!} f(PR)$$

$$Z = \sum_{p=1}^{N!} \frac{1}{N!} \int dR_1 \ldots dR_M \, e^{-\sum_{i=1}^{M} S(R_i, R_{i+1})}$$

- Means the path closes on itself with a permutation. $\mathbf{R_1 = PR_{M+1}}$
- Too many permutations to sum over; we must <u>sample</u> them.
- PIMC task: sample path { $R_1, R_2, \ldots R_M$ and P} with Metropolis Monte Carlo (MCMC) using "action", S, to accept/reject.

Exchange picture

- Average by sampling over all paths and over connections.
- Trial moves involve reconnecting paths differently.
- At the superfluid transition a “macroscopic” permutation appears.
- This is reflection of bose condensation within PIMC.



3 boson example

- Suppose the 2 particle action is exact.
- Make Jastrow approximation for spatial dependance (Feynman form)

$$\left\langle R\left|e^{-\beta H}\right|R'\right\rangle = e^{-\sum_i (r_i - r_i')^2} \prod_{i<j} f\left(r_{ij}, r_{ij}'\right) \qquad \text{units with } 4\beta\lambda = 1$$

$$\rho_{bose}(R) = \sum_P \left\langle R\left|e^{-\beta H}\right|PR\right\rangle \sim \left[\sum_P \left\langle R\left|e^{-\beta H_0}\right|PR\right\rangle\right] \prod_{i<j} f\left(r_{ij}, r_{ij}\right)$$

$$\rho_{bose}(R) = \left|\Psi(R)\right|^2 \left[1 + e^{-r_{12}^2} + e^{-r_{13}^2} + e^{-r_{23}^2} + 2e^{-r_{12}^2 - r_{23}^2 - r_{13}^2}\right]$$

- Spatial distribution gives an effective attraction (bose condensation).
- For 3 particles we can calculate the “permanent” but larger system require us to sample it.
- Anyway permutations are more physical.

Permutation Sampling

For bosons we also have to move through permutation space. A "local" move is to take an existing permutation and multiply by a k-cycle

$$P_{trial} = \hat{p}(i_1, i_2, ..., i_k)P$$

- Sometimes need more than 2-particle exchanges-for fermions 3 particle exchanges are needed
- Need more than 1 time slice because of hard core.

Two alternative ways:

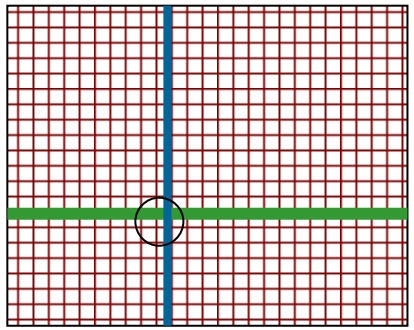
1. Make a table of possible exchanges and update the table. Good for up to 4 particle exchanges "SELECT"
2. Have a virtual table and sample permutation from that table. Good for longer exchanges (up to 10 body exchanges). "PERMUTE"

Heat Bath Method

Sample a neighborhood of a given point so that it is in local equilibrium.

$$T(s \to s') = \frac{\pi(s')}{C(s)}$$

$$C(s) = \sum_{s'' \in N(s)} \pi(s'')$$



Then the acceptance probability will be: $A(s \to s') = \min\left(1, \frac{C(s)}{C(s')}\right)$

Can only be used if it is possible to quickly compute the normalization.

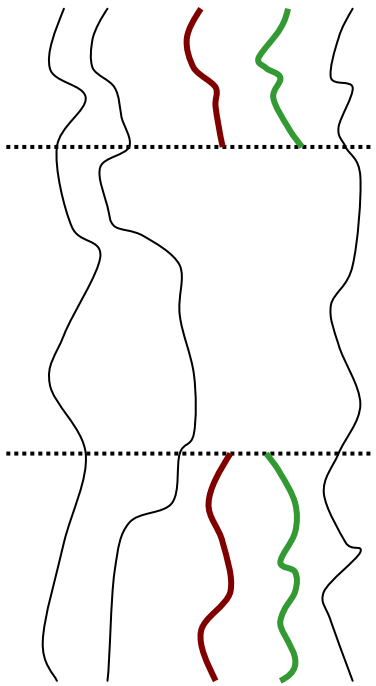
Acceptance ratio=1 if C(s) is independent of s.

For a given neighborhood, convergence is as fast as possible (it equilibrates in one step).

How to select permutation

- Heat bath probability of move being accepted is:

$$T(p) \propto \left\langle pR \middle| e^{-n\tau H} \middle| R' \right\rangle \approx \exp\left[-\frac{\sum_j \left(r_j - r_{pj}' \right)^2}{4n\lambda\tau} \right] = \prod_j h_{j,Pj}$$



$$h_{i,j} = \exp\left[-\frac{\left(r_j - r_i' \right)^2}{4n\lambda\tau} \right] = \text{distance table}$$

Example: $T(1,2) \propto h_{1,2} h_{2,1}$

  - Set up "h" matrix
  - Loop over all pairs and find all T(i,j)'s
  - Loop over triplets and find T(I,j,k)'s ...
  - In acceptance probability we need the normalized probability: $\hat{T} = \frac{T}{C}$ where $C = \sum_p T(p)$
  - This gives an addition rejection rate.

Discrete Distributions

Any discrete distribution p_k can be sampled by constructing the cumulant.

$$c_k = \sum_{i=1}^{k} p_i$$

- Sample $0<u<1$.
- Find which region it is in. i.e. find k: $c_{k-1}<u<c_k$
- Return label "k".

- The search operation can be done by bisection in $\log_2(N)$ steps.

Bisection method

1. Select time slices

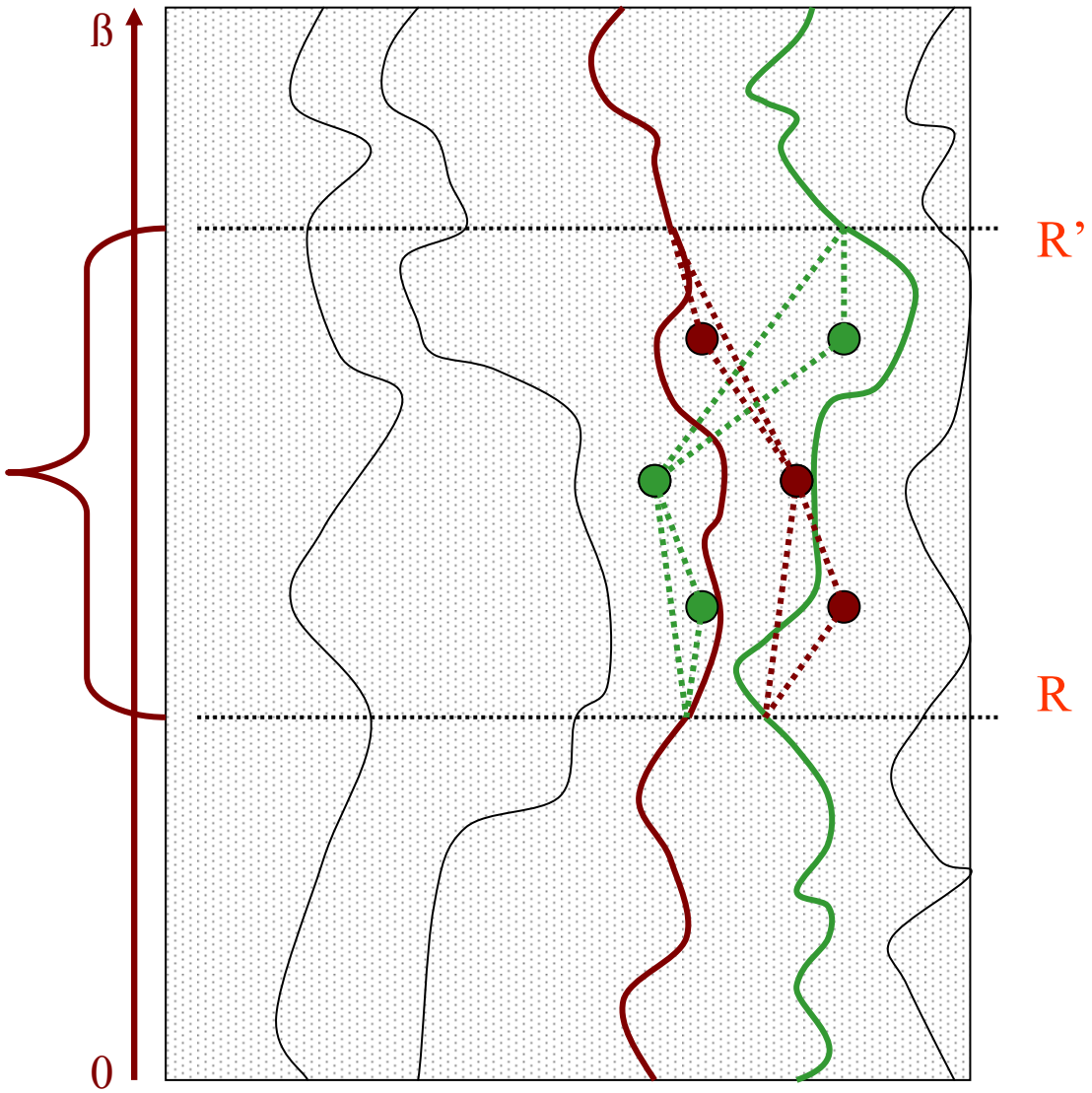
2. Select permutation from possible pairs, triplets, from:

$$\rho(R, PR'; 4\tau)$$

3. Sample midpoints

4. Bisect again, until lowest level

5. Accept or reject entire move



Liquid helium
the prototypic quantum fluid

- A helium atom is an elementary particle. A weakly interacting hard sphere. First electronic excitation is 230,000 K.
- Interatomic potential is known more accurately than any other atom because electronic excitations are so high.

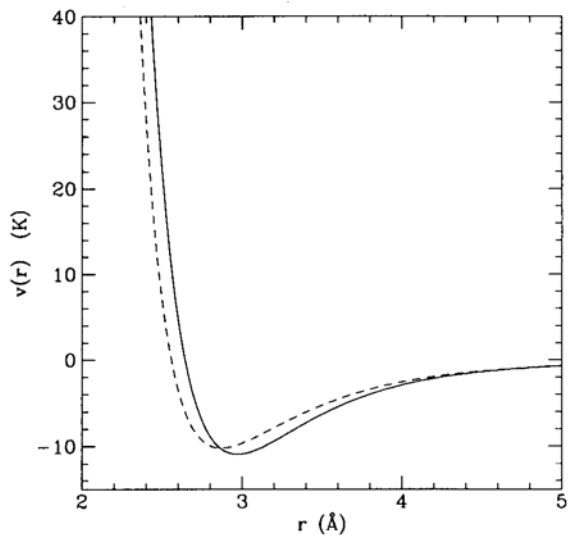


FIG. 1. The semiempirical pair potential between two helium atoms: solid line, Aziz *et al.* (1992); dashed line, Lennard-Jones 6-12 potential with $\epsilon = 10.22$ K and $\sigma = 2.556$ Å.

- Two isotopes:
  - ^{3}He (fermion: antisymmetric trial function, spin 1/2)
  - ^{4}He (boson: symmetric trial function, spin zero)

Helium phase diagram

- Because interaction is so weak helium does not crystallize at low temperatures. Quantum exchange effects are important
- Both isotopes are quantum fluids and become superfluids below a critical temperature.
- One of the goals of computer simulation is to understand these states, and see how they differ from classical liquids starting from non-relativistic Hamiltonian:

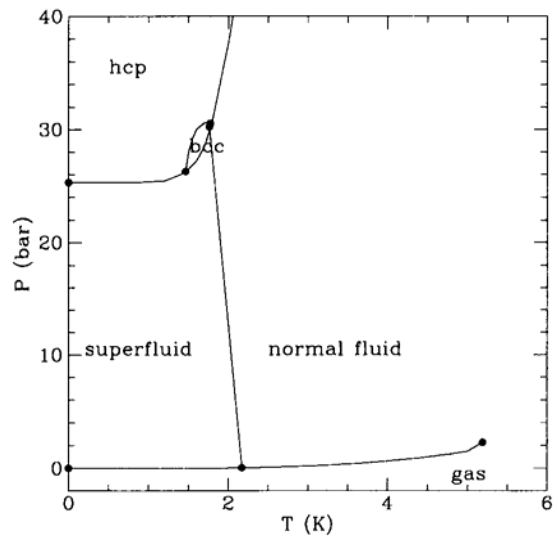


FIG. 2. The phase diagram of ^{4}He.

$$\widehat{H} = -\sum_i \frac{\hbar^2}{2m_i}\nabla_i^2 + V(R)$$

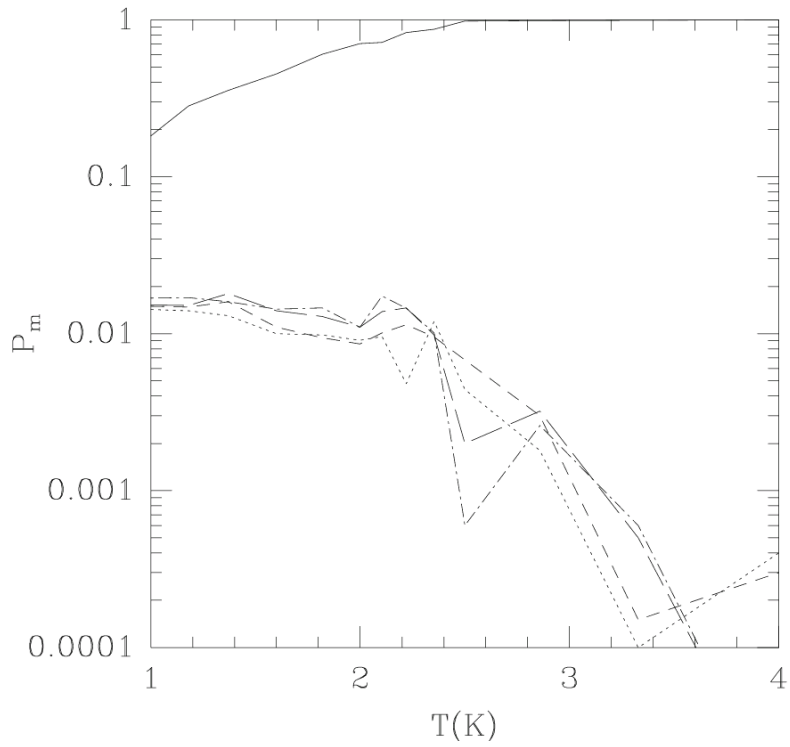
$$\lambda \equiv \frac{\hbar^2}{2m_i}$$

Path Integral explanation of Boson superfluidity

- Exchange can occur when thermal wavelength is greater than interparticle spacing $k_B T \leq h^2 \rho^{2/d} / m$
- Localization in a solid or glass can prevent exchange.
- Macroscopic exchange (long permutation cycles) is the underlying phenomena leading to:
  - Phase transition: bump in specific heat: entropy of long cycles
  - Superfluidity winding paths
  - Offdiagonal long range order--momentum condensation separation of cut ends
  - Absence of excitations (gaps)
- Some systems exhibit some but not all of these features.
- Helium is not the only superfluid. (2001 Nobel Prize for BEC)

Permutation Distribution

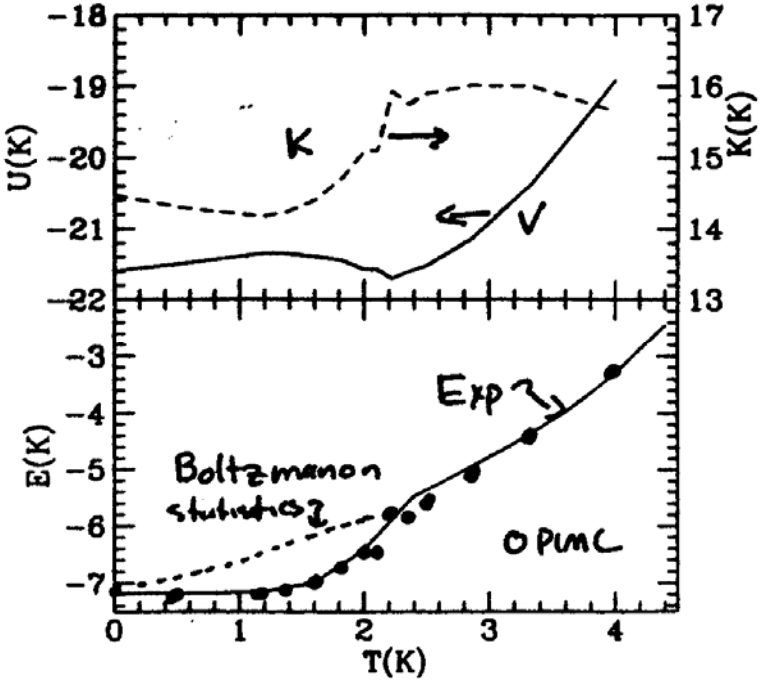
- As paths get longer probability of permutation gets significant.
- Shown is the probability of a given atom attaching itself to a permutation of length n.
- Superfluid transition occurs when there is a non-zero probability of cycle length N=size of system
- Permutations are favored in the polymer system because of entropy. In the quantum system because of kinetic energy.
- Impurities can be used to “measure” the permutations.



ENERGY

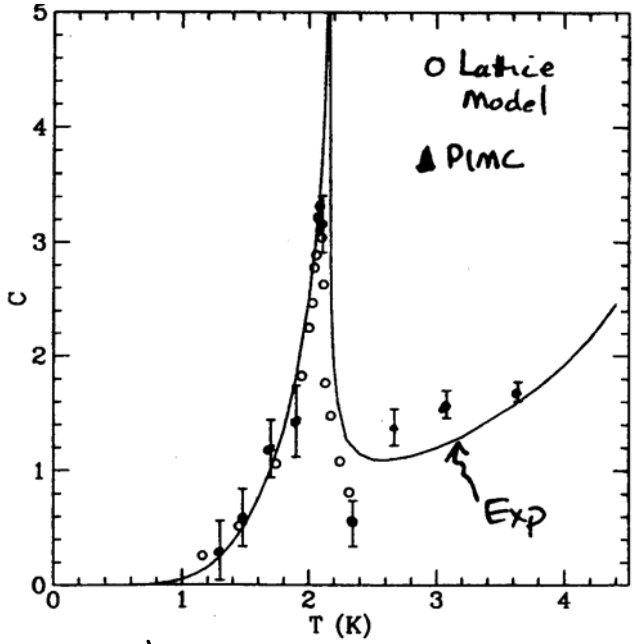
Bose statistics have a small effect on the energy

Below 1.5K ^{4}He is in the ground state.



SPECIFIC HEAT

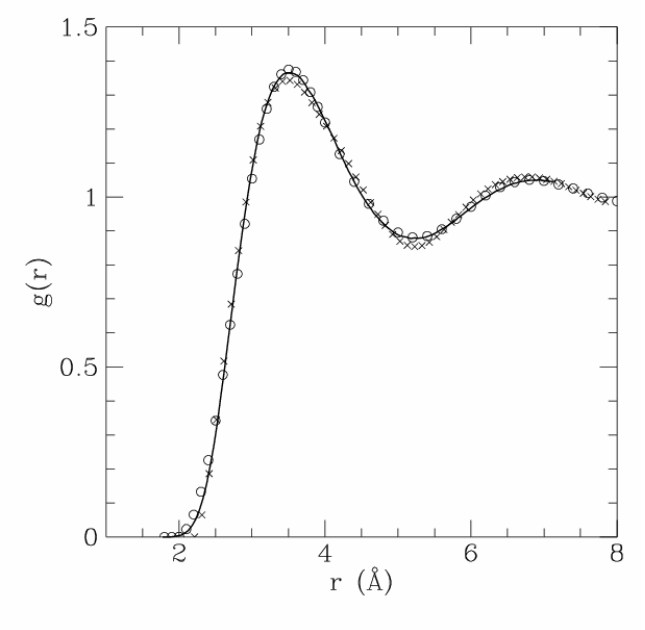
- Characteristic λ shape when permutations become macroscopic
- Finite size effects cause rounding above transition

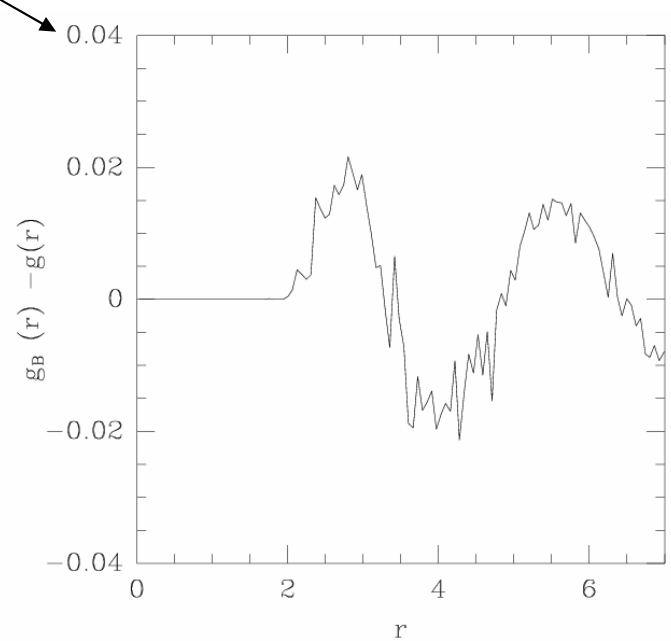


$$E = \left\langle V + \frac{3 N_{cycles}}{2\beta} + \frac{1}{2}(r_i - c_i)\cdot\nabla_i V \right\rangle$$

Kinetic term becomes smaller because $N_{cycle} < N$. Springs stretched more.

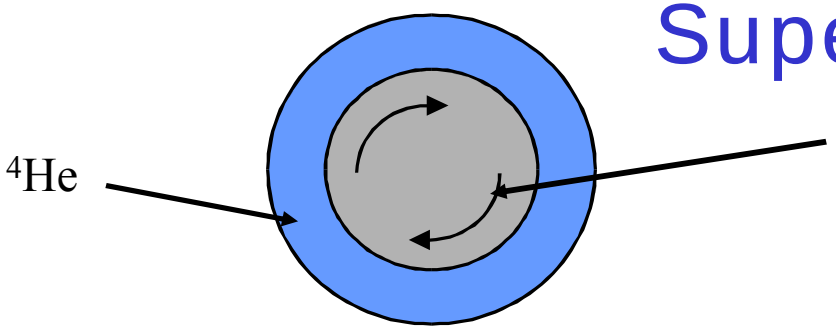
- Transition is not in the static distribution functions like S(k) or g(r). They do not change much at the transition. NON-CLASSICAL TRANSITION
- Effect of turning off bose statistics at the transition:





- Transition is in the imaginary-time connections of the paths-the formation of the macroscopic exchange.

Superfluidity and PIMC



rotating disks:

Andronikashvili's expt (1946)

$$(\rho_s + \rho_N \equiv \rho)$$

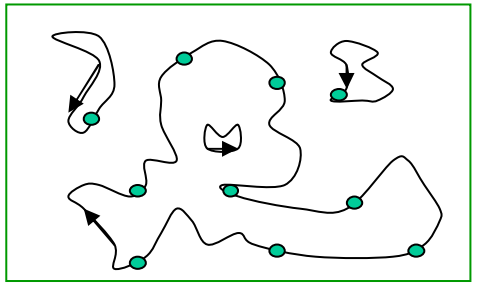
- We define superfluidity as a linear response to a velocity perturbation (the energy to rotate the system) Landau definition.

$$\frac{\rho_s}{\rho} = 1 - \frac{I}{I_c} = \left.\frac{dF}{d\omega^2}\right|_{\omega=0}$$

- To evaluate with Path Integrals, we use the Hamiltonian in rotating frame:

$$\hat{H}_\omega = \hat{H}_0 - \omega \hat{L}_z$$

$$\frac{\rho_s}{\rho} = 1 - \frac{1}{I_c}\left\langle \int_0^\beta dt\, \hat{L}_z e^{-(\beta-t)\hat{H}_0} \hat{L}_z e^{-t\hat{H}_0} \right\rangle$$

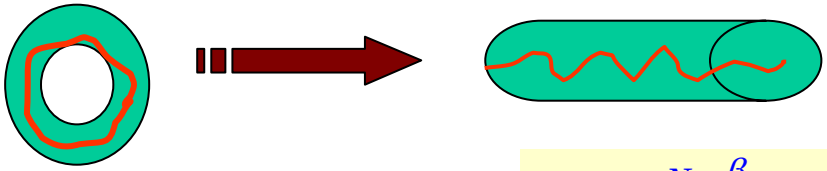


$$\frac{\rho_s}{\rho} = \frac{2m}{\beta\lambda I_c}\left\langle A_z^2 \right\rangle$$

A = signed area of imaginary-time paths

Winding numbers in periodic boundary conditions

- Distort annulus



- The area becomes the **winding** (average center of mass velocity)

$$W = \sum_{i=1}^{N} \int_0^{\beta} dt \frac{dr_i(t)}{dt}$$

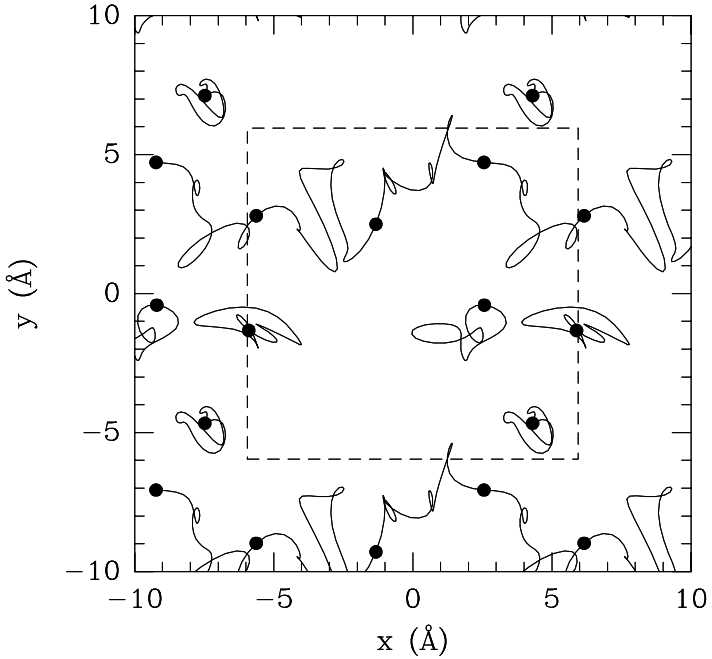
- The superfluid density is now estimated as:

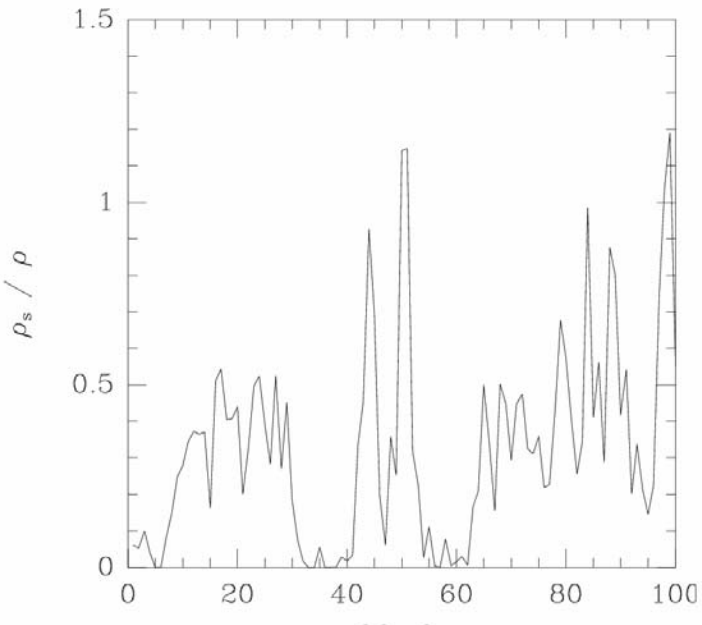
$$\frac{\rho_s}{\rho} = \frac{\langle W^2 \rangle}{2\lambda\beta N}$$

- Exact linear response formula. (analogous to relation between $\chi \sim \langle M^2 \rangle$ for Ising model.
- Relates **topological** property of paths to dynamical response. Explains why superfluid is "protected."
- Imaginary time dynamics is related to real time response.
- How the paths are connected is more important than static correlations.

Ergodicity of Winding Number

- Because winding number is topological, it can only be changed by a move stretching all the way across the box.
- For cubic boundary conditions we need $m \geq N^{1/3}$
- Problem to study finite size scaling: we get stuck in a given winding number sector.
- Advanced algorithms needed such as worm or directed loops (developed on the lattice).





Superfluidity in pure Droplets

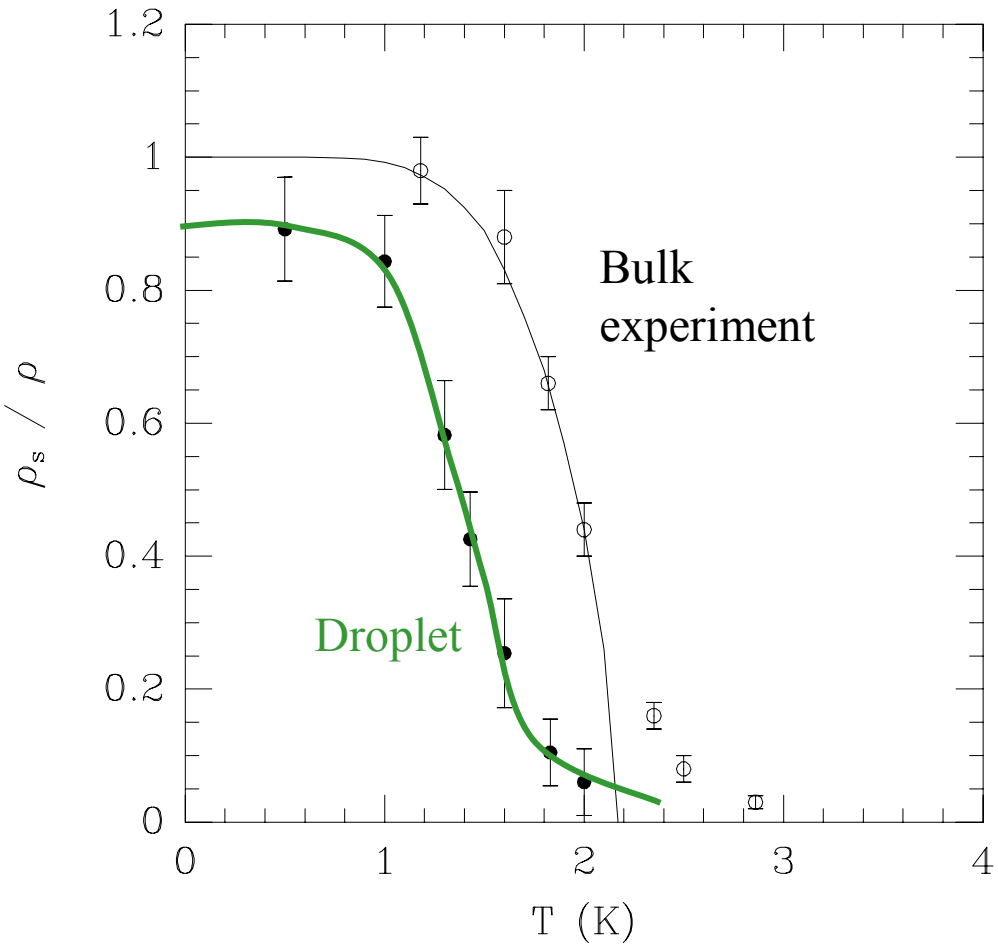
$$\frac{\rho_s}{\rho} = \frac{2m}{\beta} \frac{\langle A_z^2 \rangle}{\lambda \; I_c}$$

- 64 atom droplet goes into the superfluid state in temperature range 1K <T <2K.

  NOT A PHASE TRANSITION!

- But almost completely superfluid at 0.4K (according to response criteria.)
- Superfluidity of small droplets recently verified.

Sindzingre et al 1990



Determination of T_c

Runge and Pollock PRB.

At long wavelength, the free energy is given by the functional:

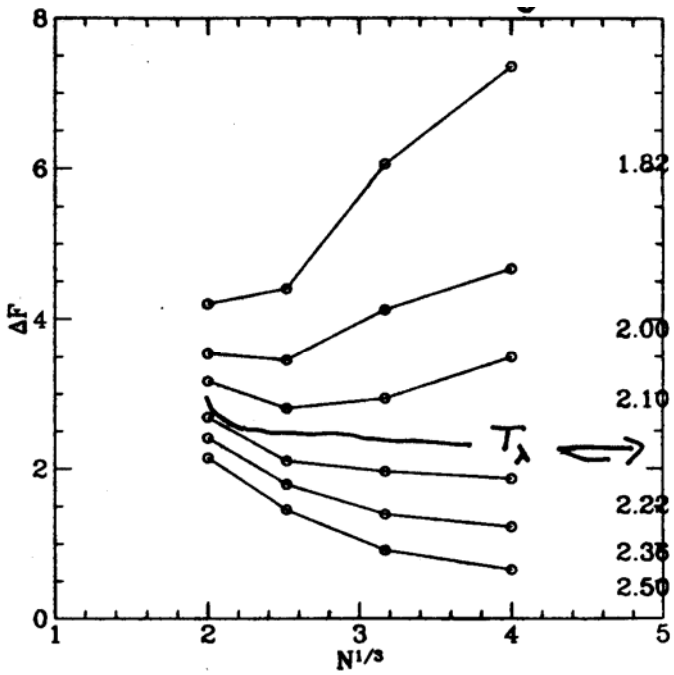
$$F = \int dr \left[\lambda \left| \nabla \phi(r) \right|^2 + \mu \left| \phi(r) \right|^2 + V \phi(r)^4 \right]$$

The energy to go from PBC to ABC is given by:

$$F = \frac{\mu \lambda \pi^2 L^{d-2}}{V} = -k_B T \left\langle e^{iW\pi/L} \right\rangle$$

We determine T_c by where F is constant with respect to number of atoms N.

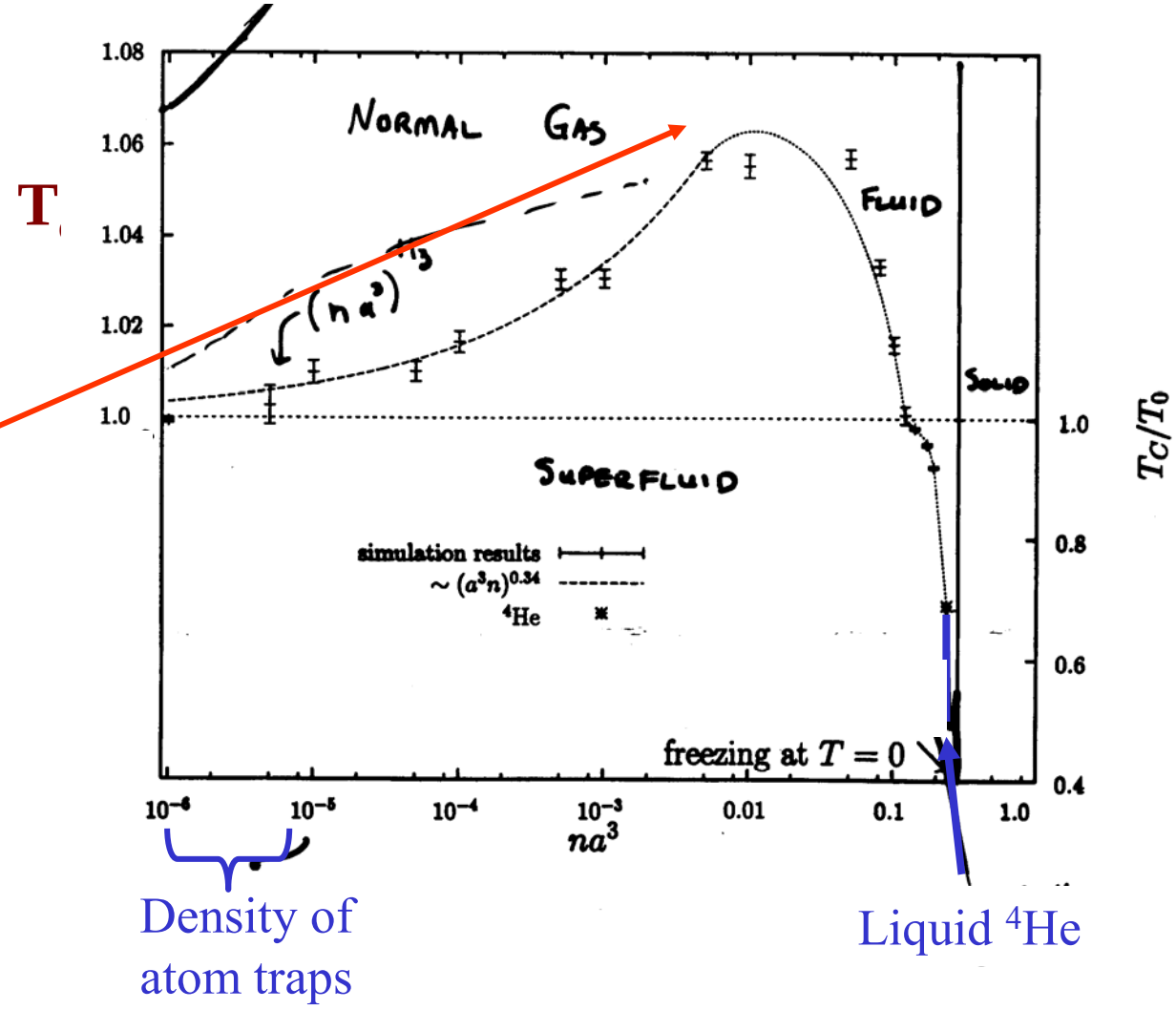
For N~100 , T_c correct to ~1%.



Phase Diagram of Hard Sphere Bosons

- Atomic traps are at low density
- With PIMC we mapped out range of densities
- There is an enhancement of T_c by 6% because of density homogenization.

Gruter et al PRL 99.



T_c

$T_c / T_{c0} \sim 1 + n^{1/3} a$

Density of atom traps

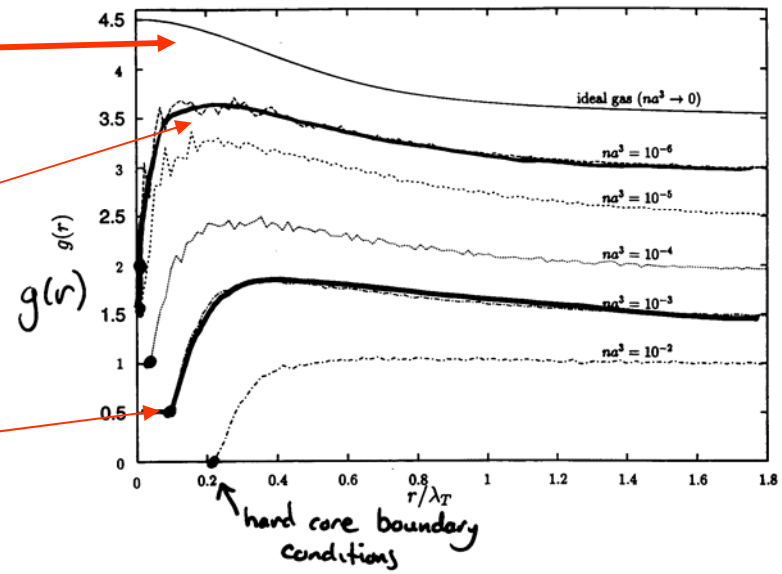
Liquid ^{4}He

Pair correlations at low density

Free boson pair correlation.

Peak caused by attraction of bosonic exchange.

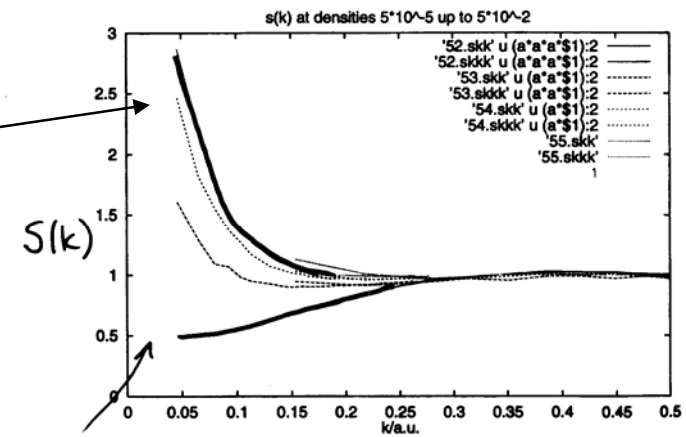
Zero is hard core interaction.



Structure Factor S(K)

(FT of density)

Large compressiblity near T_c (clustering)



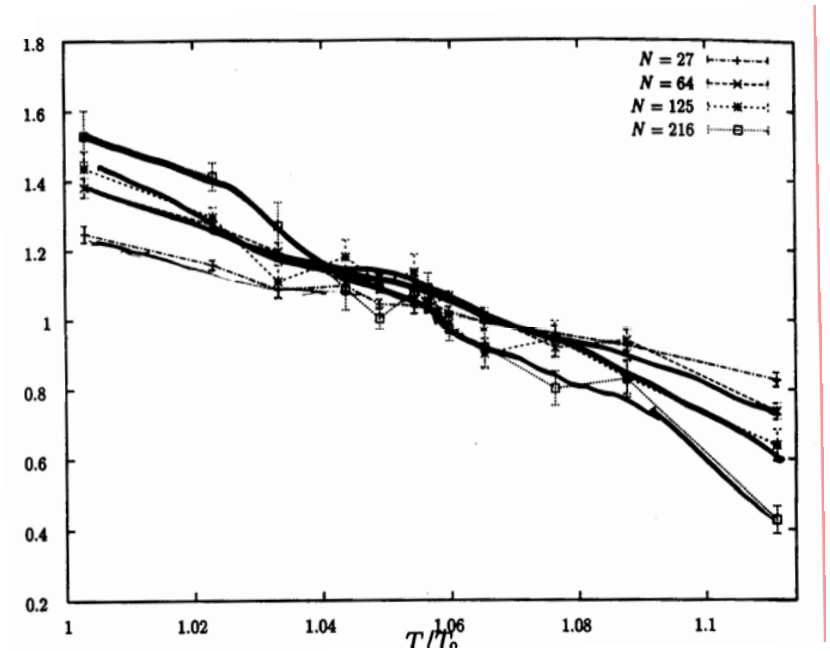
Determination of T_c using superfluid density. Finite size scaling.

$$\rho_s = \frac{1}{L} Q\left(\tfrac{L}{\zeta}\right)$$

$$\zeta = \text{bulk correlation length}$$

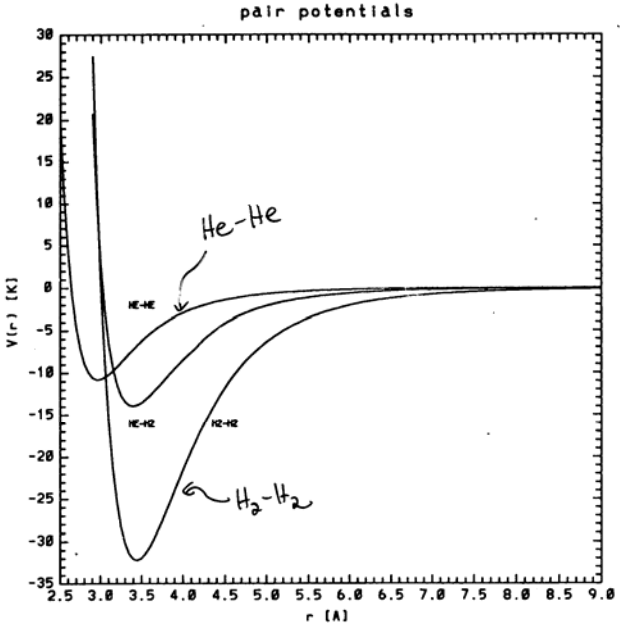
$$\zeta \to \left(\frac{T - T_c}{T_c}\right)^{-\nu}$$

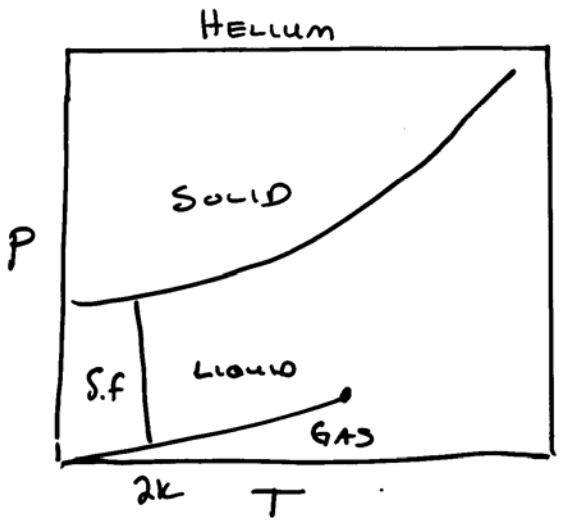
- Near T_c a single length enters into the order parameter.
- Write superfluid density in terms of the available length.
- Determine when the curves cross to get T_c and exponent.
- Exponent is known or can be computed.

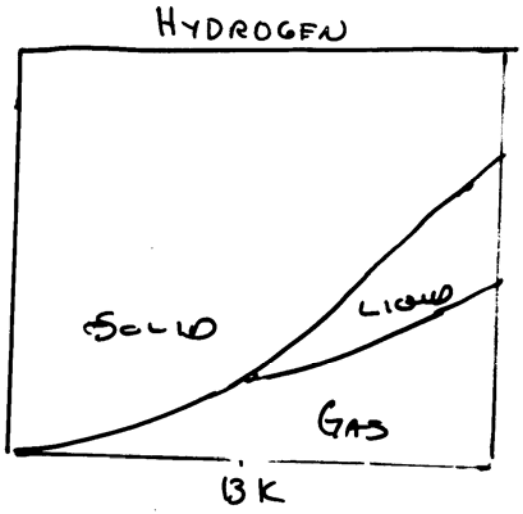


Why is H_2 not a superfluid?

- H_2 is a spherically symmetric boson like He.
- However its intermolecular attraction is three times larger
- Hence its equilibrium density is 25% higher $\Rightarrow$ solid at T<13K.
- To be superfluid we need to keep the density lower or frustrate the solid structure.





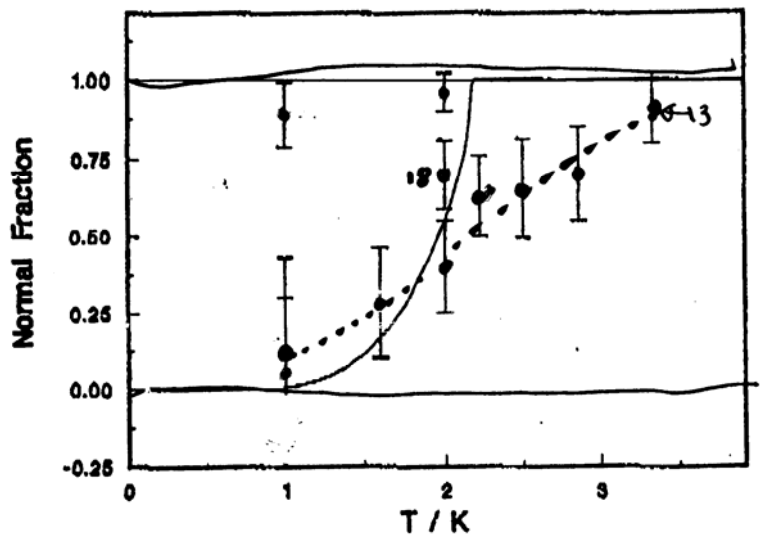


At low T and density, orientational energies are high$\Rightarrow H_2$is spherical.

$$E(J_1) - E(J_0) = 170K$$
$$E(J_2) - E(J_0) = 510K$$

H_2 droplets

- In droplet or at surfaces, many bonds are broken.
- We found that small droplets are superfluid.
- Recently verified in experiments of ^4He-H_2-OCS clusters: When a complete ring of H_2 surrounds OCS impurity, it no longer acts as a rigid body, but decouples from the motion of the OCS.

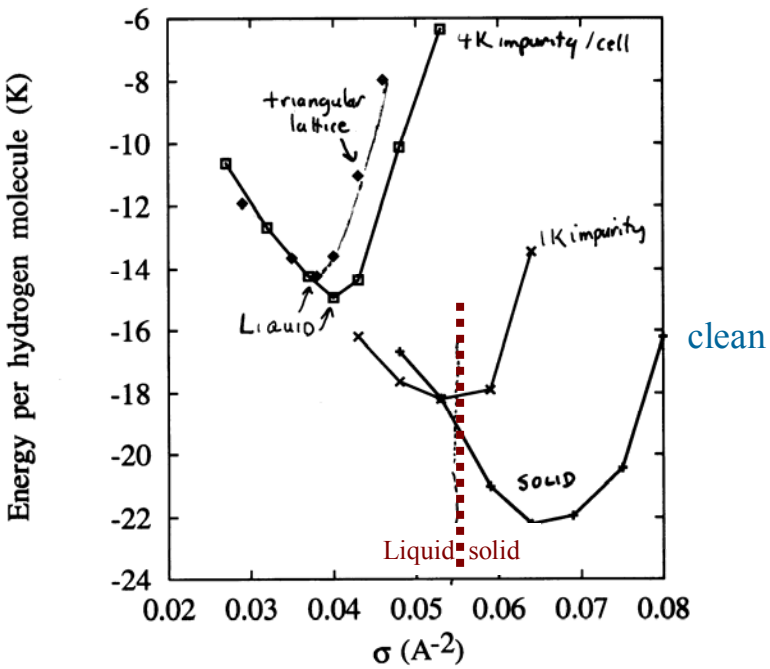


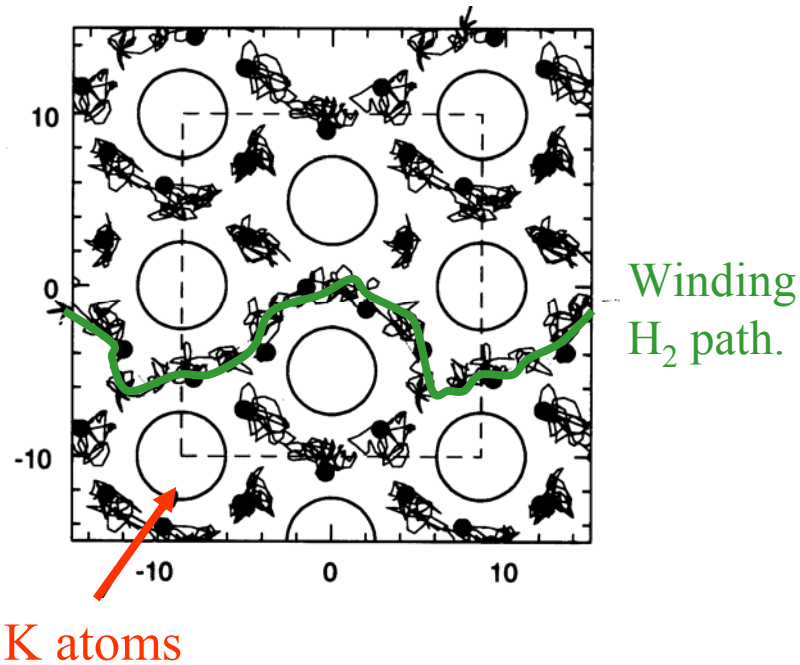
Sindzingre et al. PRL 67, 1871 (1991).

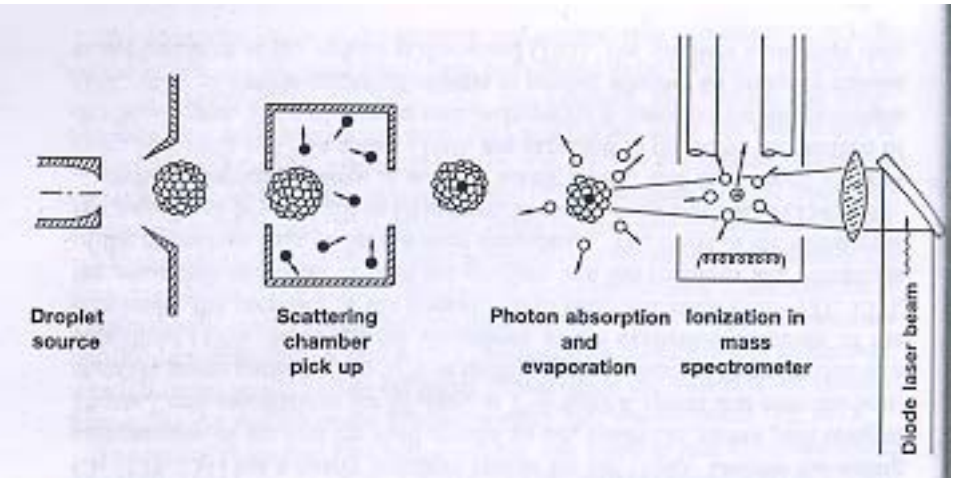
H_2 on Ag-K surfaces

Gordillo, DMC PRL 79, 3010, 1997.

- Formation of solid H_2 is frustrated by alkali metal atoms.
- Lowers the wetting density-result is liquid (superfluid) ground state with up to 1/2 layer participating.
- Has not yet been seen experimentally.







Experimental Setup for He droplets

Toennies and Vilesov,
Ann. Rev. Phys. Chem. 49, 1 (1998)

- Adiabatic expansion cools helium to below the critical point, forming droplets.
- Droplets then cool by evaporation to:

  T=0.38 K, $N \approx 10^4$ (^{4}He)

  T=0.15 K, $N \approx 10^3$ (^{3}He)
- The droplets are sent through a scattering chamber to pick up impurities, and are detected either with a mass spectrometer with electron-impact ionizer or a bolometer.
- Spectroscopy yields the rotational-vibrational spectrum for the impurity to accuracy of 0.01/cm. Almost free rotation in superfluid helium but increase of MOI of rotating impurities.

Droplets and PIMC

E. Draeger (LLNL) D. Ceperley(UIUC)

- Provide precise microscopic probes for phenomenon such as superfluidity and vortices.
- Provide a nearly ideal “spectroscopic matrix” for studying molecular species which may be unstable or weakly interacting in the gas phase.
- PIMC can be used to simulate ^{4}He droplets of up to 1000 atoms, at finite temperatures containing impurities, calculating the density distributions, shape deformations and superfluid density.
- Droplets are well-suited to take advantage of the strengths of PIMC:
  - Finite temperature (T=0.38 K)
  - Bose statistics (no sign problem)
  - Finite size effects are interesting.

Demonstration of droplet superfluidity

Grebenev, Toennies, Vilesov: *Science 279, 2083 (1998)*

- An OCS molecule in a ^{4}He droplet shows rotational bands corresponding to free rotation, with an increased moment of inertia (2.7 times higher)

  → ^{4}He are "coat" the impurity allowing it to freely rotate in the superfluid

- They replaced boson ^{4}He with fermion ^{3}He. If Bose statistics are important, then rotational bands should disappear. → they didn't!

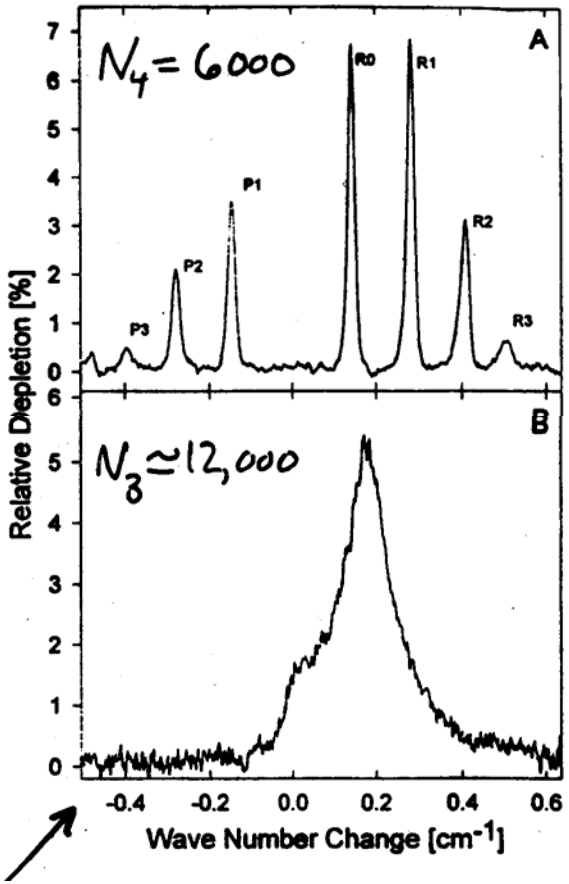
- However, commercial ^{3}He has ^{4}He impurities, which would be more strongly attracted to an impurity.

  → How much ^{4}He does it take to "coat" the impurity and get free rotation?

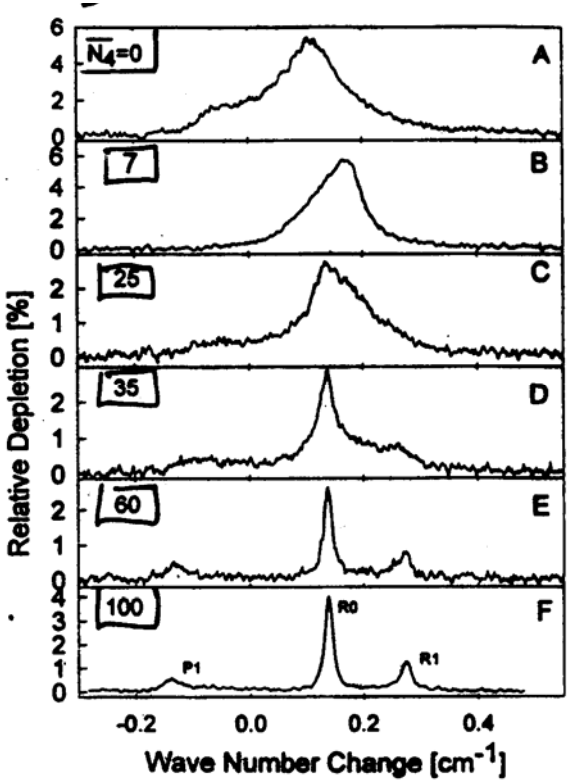
- They found that it takes around 60 ^{4}He atoms.

^{4}He

^{3}He



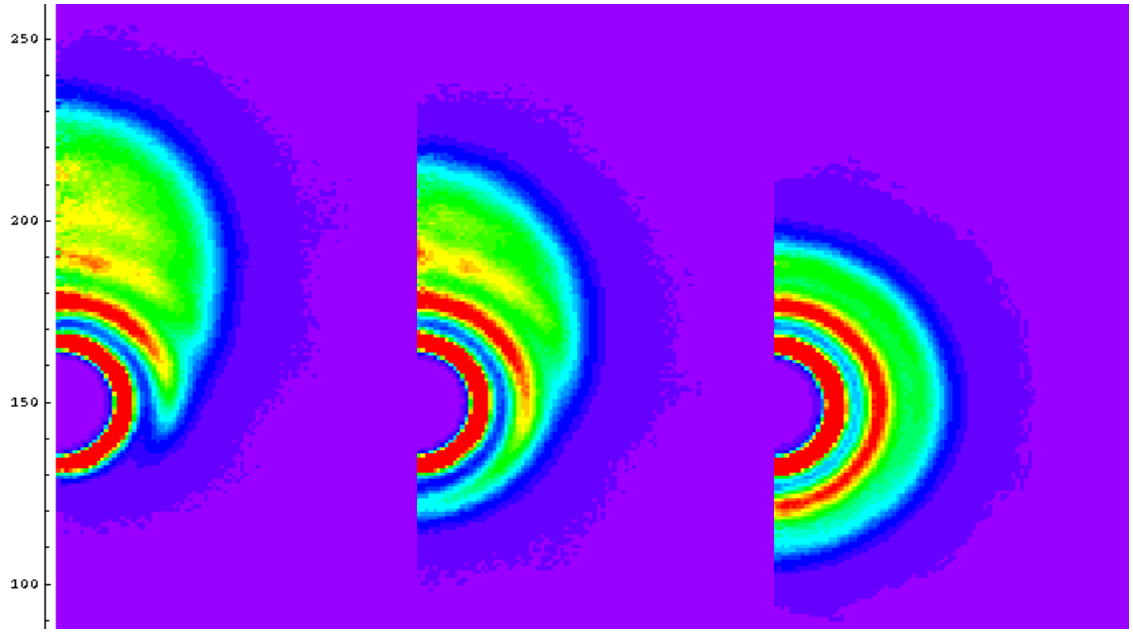
Small impurities of ^{4}He in ^{3}He



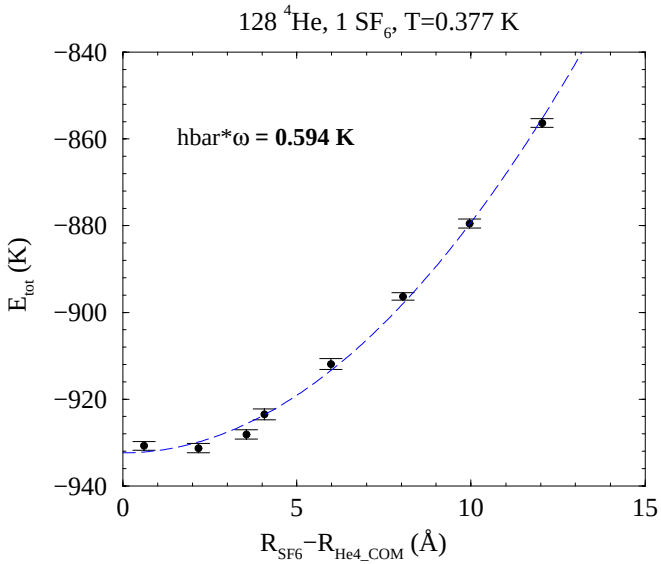
^{4}He is more strongly attracted to impurity because of zero point effects, so it coats the impurity, insulating it from the ^{3}He.

Density distribution within a droplet

- Helium forms shells around impurity (SF_6)
- During addition of molecule, it travels from the surface to the interior boiling off 10-20 atoms.



- How localized is it at the center?
- We get good agreement with experiment using the energy vs. separation from center of mass.



Local Superfluid Density Estimator

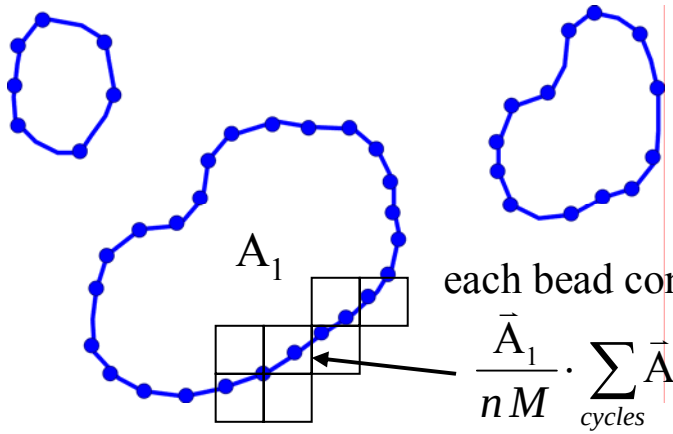
Although superfluid response is a non-local property, we can calculate the local contribution to the total response.

$$\rho_s = \frac{4m^2\rho}{3\hbar^2\beta I_c\Omega^2}\int d\vec{r}\, d\vec{r}'\, \vec{A}(\vec{r})\cdot\vec{A}(\vec{r}') = \frac{1}{\Omega}\int d\vec{r}\, \rho_s(\vec{r})$$

Where A is the area.

$$\vec{A}(\vec{r}) \equiv \int_0^\beta dt \sum_i (\vec{r}_{it}\times\vec{v}_{it})\,\delta(\vec{r}_{it}-\vec{r})$$

$$\Rightarrow \quad \rho_s(\vec{r}) = \frac{4m^2 N}{3\hbar^2\beta I_c}\int d\vec{r}'\, \vec{A}(\vec{r})\cdot\vec{A}(\vec{r}')$$



each bead contributes $\frac{\vec{A}_1}{nM}\cdot\sum_{cycles}\vec{A}_c$

$$\rho_s(\vec{r}) \propto \frac{\vec{A}_1}{n\ \ M}\cdot\sum_{cycles}\vec{A}_c = \frac{A_1^2}{n\ \ M} + \frac{\vec{A}_1}{n\ \ M}\cdot\sum_{c\neq 1}\vec{A}_c$$

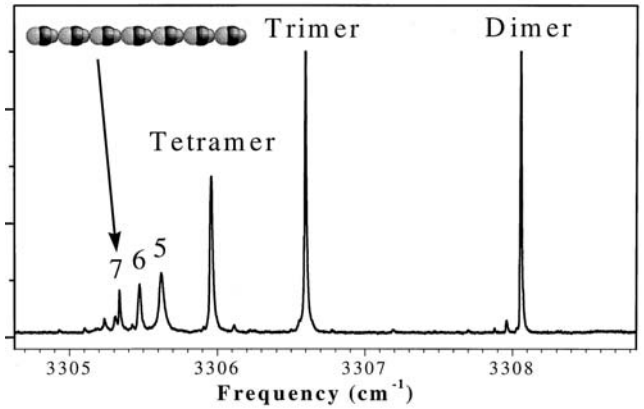
approximation: use only diagonal terms

not positive definite could be noisy

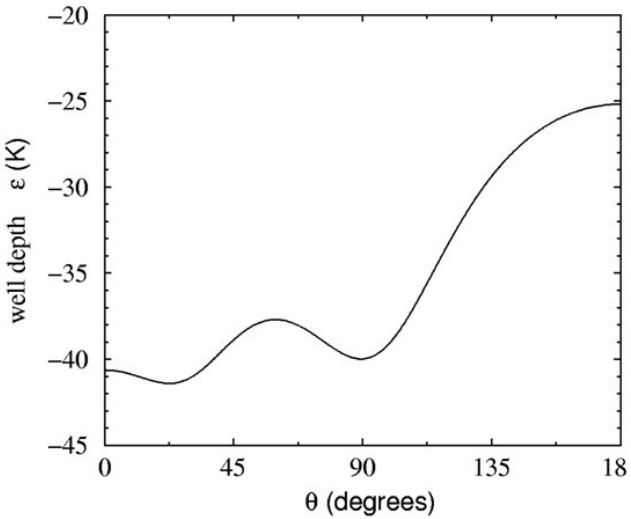
$(HCN)_x$: Self-Assembled Linear Isomers

Nauta and Miller: HCN molecules in ^{4}He droplets self-assemble into linear chains

- They measured the rotational constants for $(HCN)_1$, $(HCN)_2$, and $(HCN)_3$.
- Adiabatic following holds for $(HCN)_3$, allowing us to compare both models to experiment.
- Line vortices are unstable in pure helium droplets. Linear impurity chains may stablize and pin them.



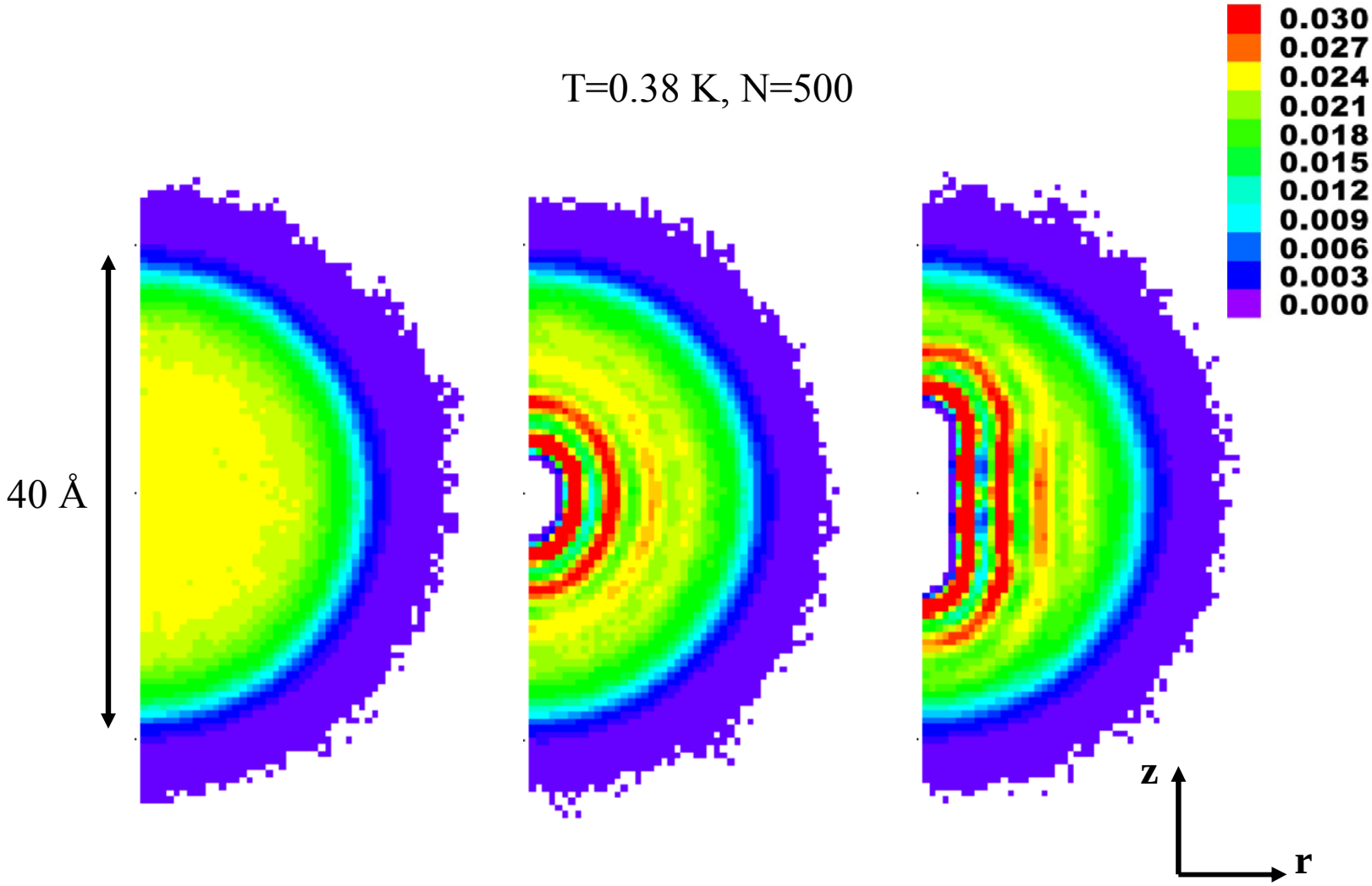
K. Nauta, R. E. Miller, Science **238**, 1895 (1999).



Atkins and Hutson: Calculated the anisotropic ^{4}He-HCN pair potential from experimental scattering data. This fit can be reproduced within error bars by a sum of three spherical Lennard-Jones potentials and a small anisotropic term.

Density Distribution of $^4He+(HCN)_x$ Droplets

T=0.38 K, N=500



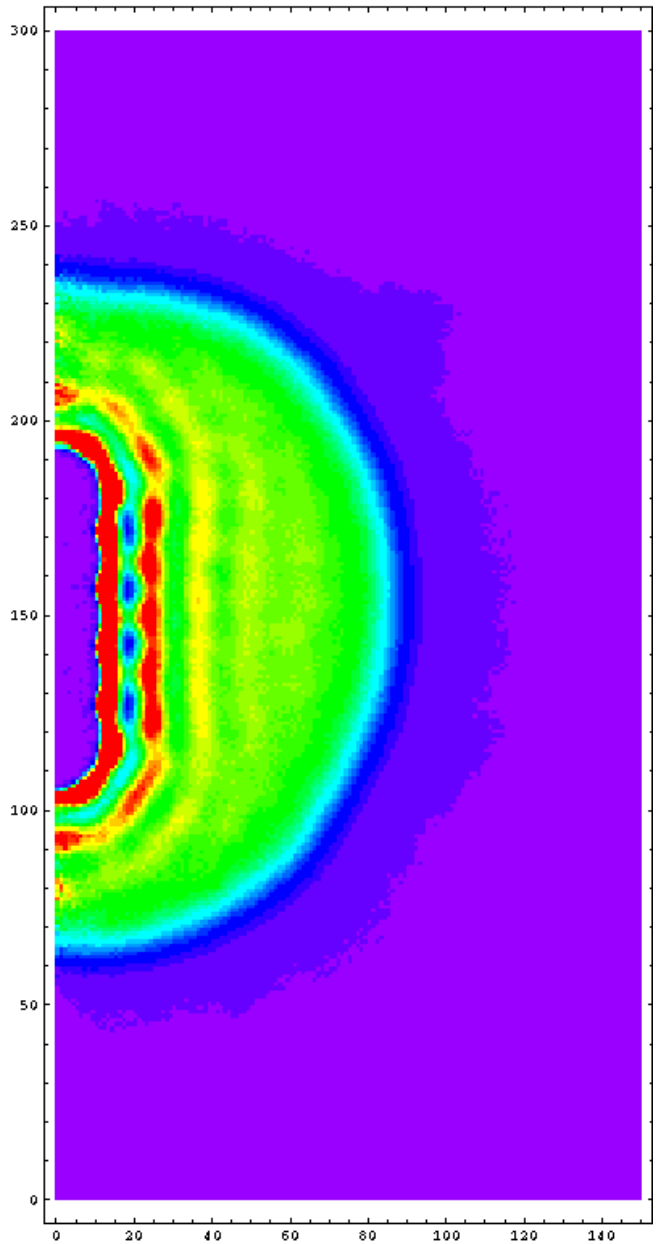
pure 1 HCN 3 HCN

Superfluidity around a linear molecule

Draeger + DMC

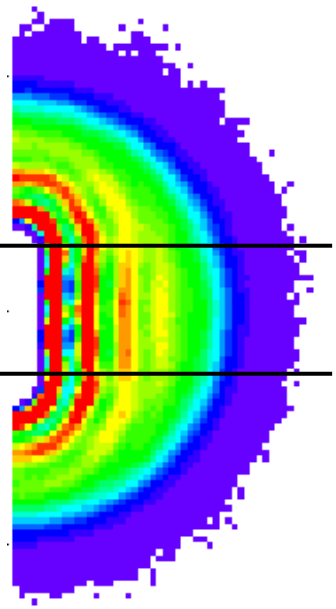
- K. Nauta and R.E. Miller (Science **283**, 1895 (1999)) found that HCN molecules will line up in a linear chain in a helium droplet, and measured the HCN-HCN spacing.
- In vacuum they form rings!
- Systems with up to 10 molecules observed.
- Use area formula to find superfluid response.
- We find almost complete superfluid system, even near the impurity.

$(HCN)_{10}$ $(^4He)_{1000}$

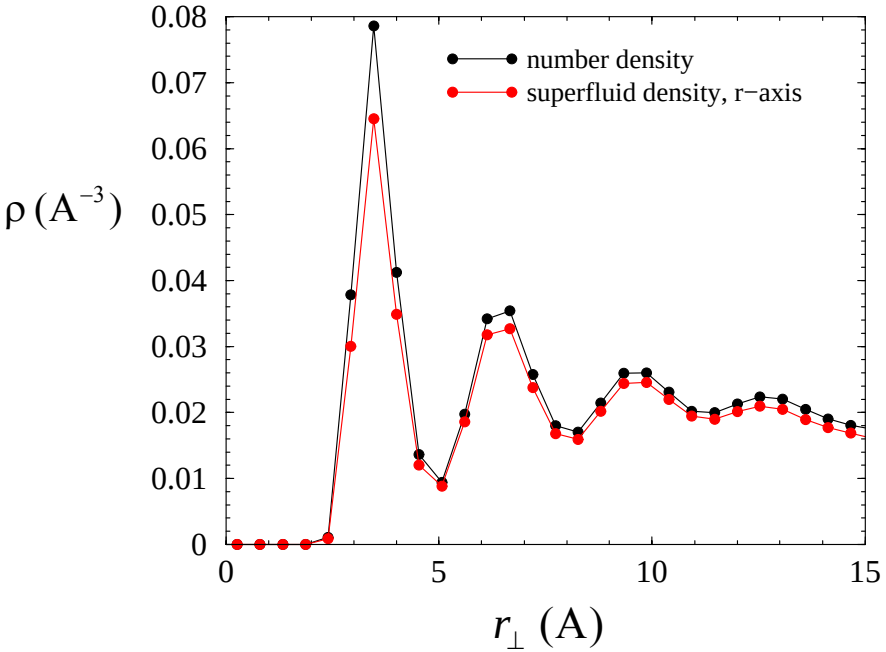


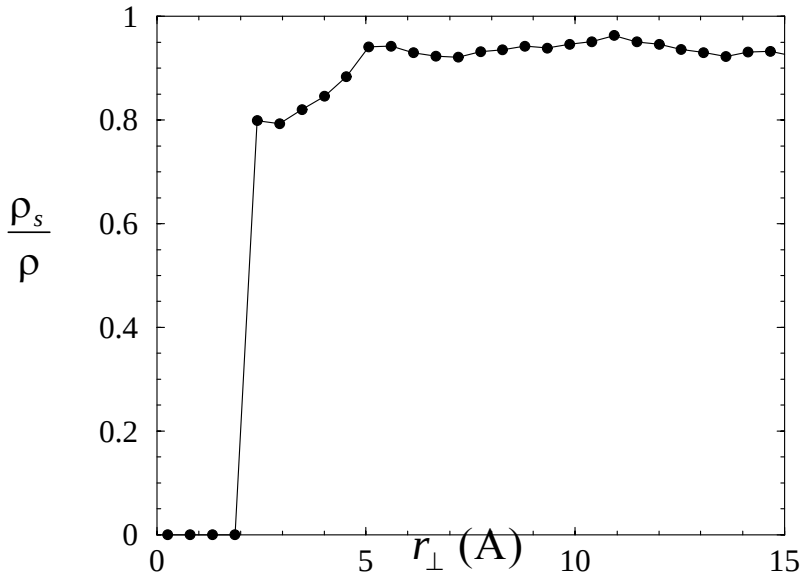
Local Superfluid Reduction

Average over region with cylindrical symmetry



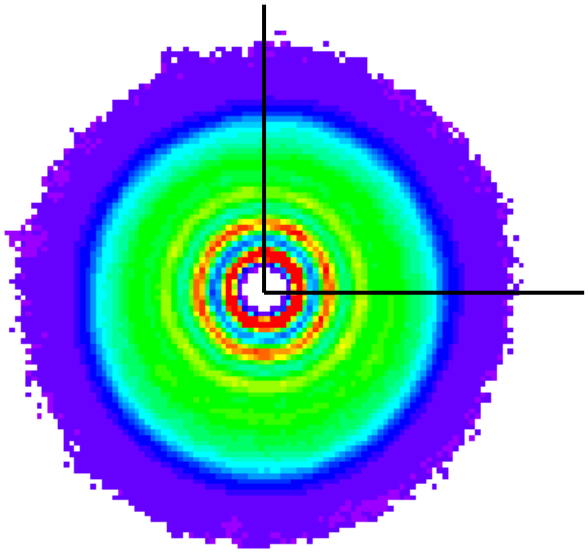
T=0.38 K, N=500
$(HCN)_3$

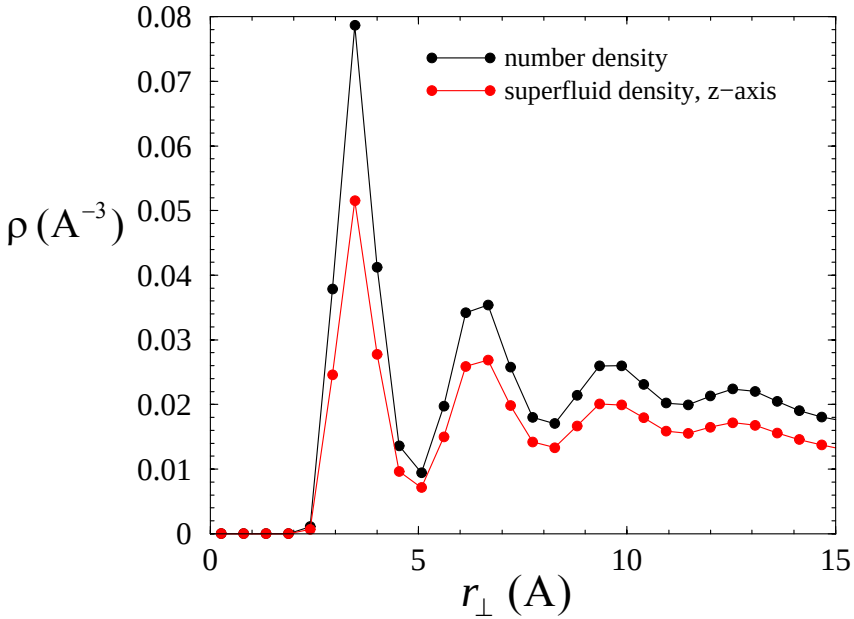




The local superfluid estimator shows a decrease in the superfluid response throughout the first solvation layer.

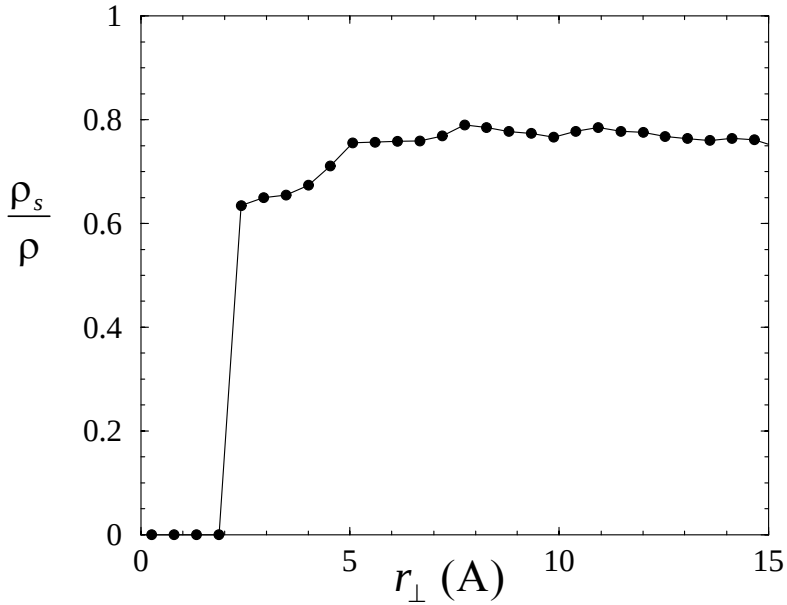
Local Superfluid Reduction





Hydrodynamic model prediction: rotating the molecule about its symmetry axis will produce no change in the free energy.

Our local superfluid calculations show a clearly defined decrease in the superfluid density in the first layer!



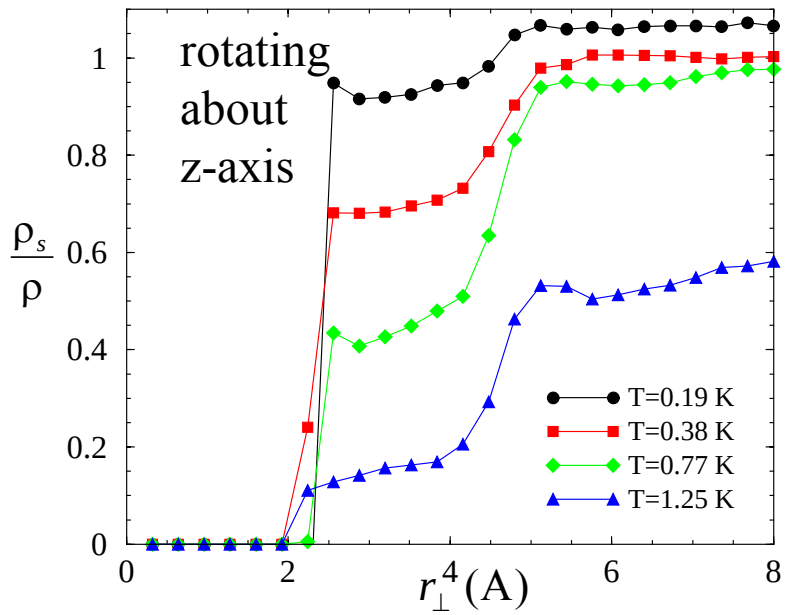
Local Superfluid Density vs. Temperature

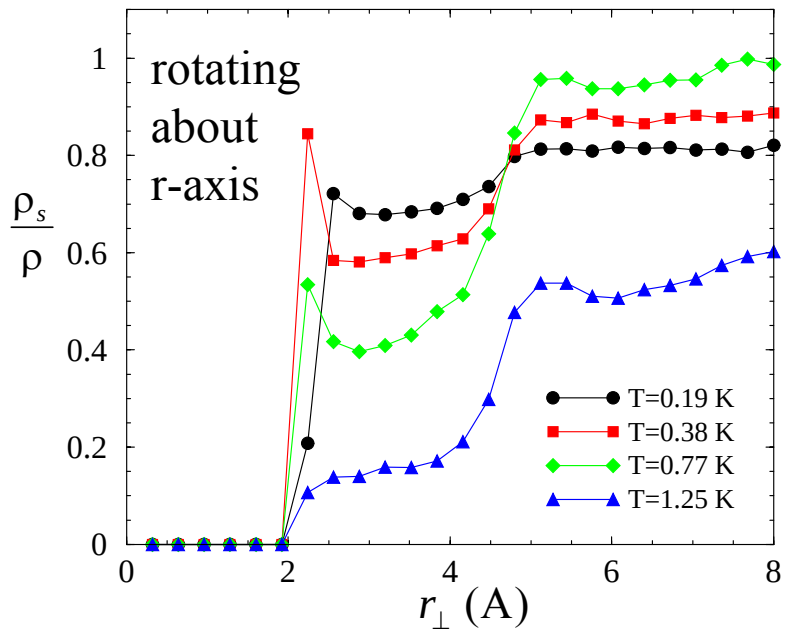
We calculated the superfluid density distribution of N=128 ^{4}He droplets with an $(HCN)_3$ isomer at several temperatures

The superfluid density in the first layer is temperature dependent!

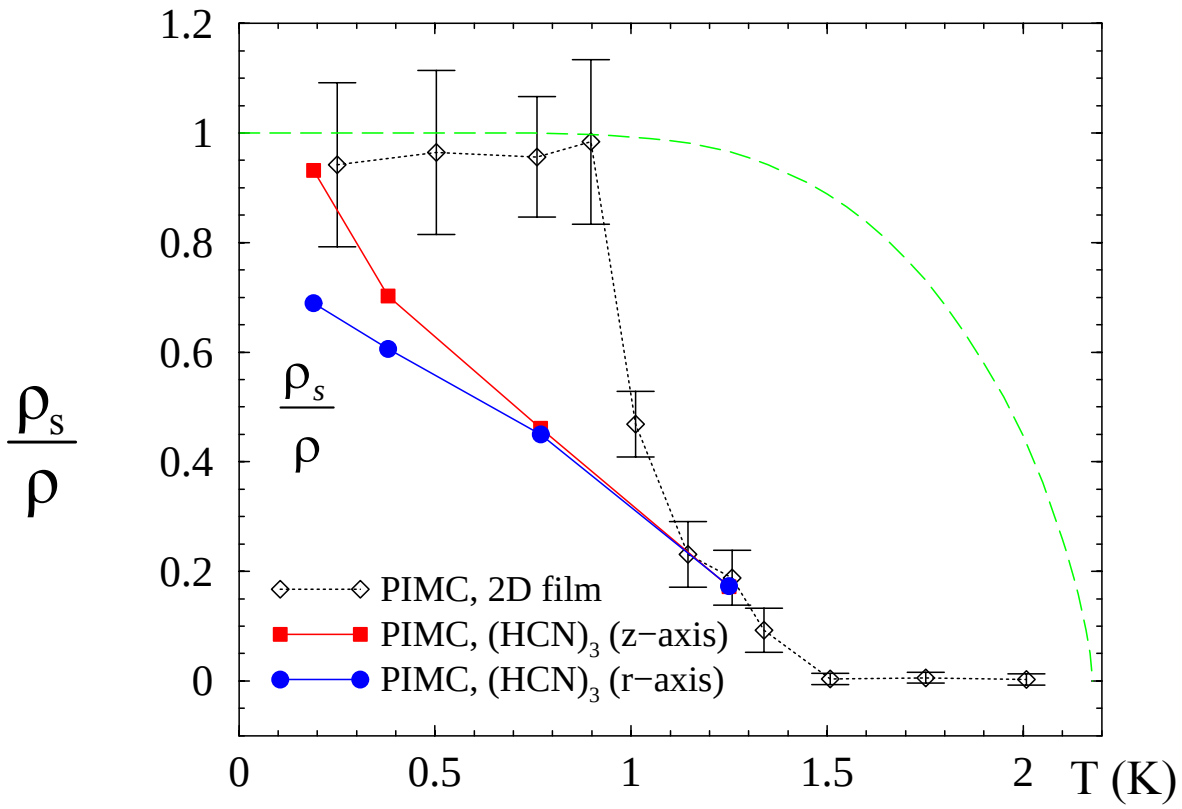
Bulk ^{4}He is 100% superfluid below 1.0 K. Both experimental measurements on helium films and PIMC studies of 2D helium show transition temperatures T_c which are significantly lower than bulk helium.

The first layer is a two-dimensional system with important thermal excitations at 0.4K: "vortex-antivortex excitations".





First Layer Superfluid Density vs. T



M. C. Gordillo and D. M. Ceperley, Phys. Rev. B **58**, 6447 (1998)

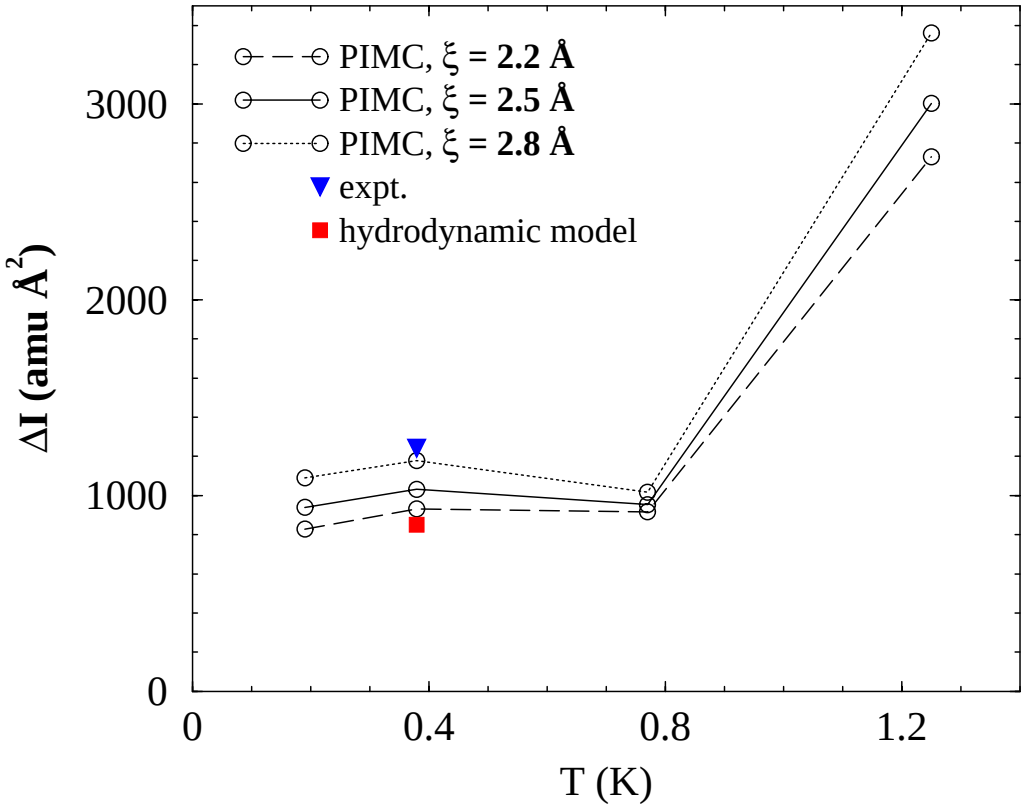
- Very broad transition, due to the small number of atoms in first layer (around 30)
- How will this affect the moment of inertia?

Moment of Inertia

The moment of inertia due to the normal helium does not depend on temperature below 1.0 K.

This is in agreement with experimental results, which found that the moment of inertia of an OCS molecule was the same at T=0.15 K and T=0.38 K.

We only looked at the superfluid density in the cylindrically-symmetric region of the first layer, not the entire first layer.



$$\Delta I = \int_{v(\xi)} d\vec{r}\, m_4 r_\perp^2 \left(\rho(\vec{r}) - \rho_s(\vec{r})\right)$$

- We have calculated the condensate fraction and density-density correlation functions throughout the free surface. We find that the surface is well-represented as a dilute Bose gas, with a small ripplon contribution.

- We have derived a local superfluid estimator, and directly calculated the normal response of helium droplets doped with HCN molecules.

- We find that the helium in the first solvation layer is a two-dimensional system, with a thermal excitations at T=0.38 K. Explains observation of Q-branch.

- The moment of inertia due to the normal fluid is dominated by the contribution from helium at the ends of the linear molecule, which is independent of temperature below T=1.0 K.

Bose condensation

- BEC is the macroscopic occupation of a single quantum state (e.g. momentum distribution in the bulk liquid).

$$n_k = \int \frac{d^3 r d^3 s}{(2\pi)^3 V} \exp(-ik(r-s)) n(r,s)$$

- The one particle density matrix is defined in terms of open paths:

$$n(r,s) = \frac{V}{Q} \int dr_2 \ldots dr_N \left\langle r, r_2 \ldots r_N \middle| e^{-\beta H} \middle| s, r_2 \ldots r_N \right\rangle$$

- We cannot calculate n(r,s) on the diagonal. We need one open path, which can then exchange with others.
- Condensate fraction is probability of the ends being widely separated versus localized. **ODLRO** (off-diagonal long range order) (*The FT of a constant is a delta function.)*
- The condensate fraction gives the linear response of the system to another superfluid.

Derivation of momentum formula

- Suppose we want the probability n_k that a given atom has momentum hk.
- Find wavefunction in momentum space by FT wrt all the coordinates and integrating out all but one atom

$$\Pr(k_1,..k_N) = \left| \int dR\ e^{-i(k_1 r_1 + ... + k_N r_N)} \Psi(R) \right|^2$$

$$n_k = \int dk_2 .... dk_N \Pr(k, k_2, ... k_N)$$

- Expanding out the square and performing the integrals we get.

$$n_k = \int \frac{d^3 r d^3 s}{(2\pi)^3 V} \exp(-ik(r-s)) n(r,s)$$

Where: $$n(r,s) = \frac{V}{Q} \int dr_2 ... dr_N \left\langle r, r_2 ... r_N \middle| e^{-\beta H} \middle| s, r_2 ... r_N \right\rangle$$

occupy the states with the Boltzmann distribution.

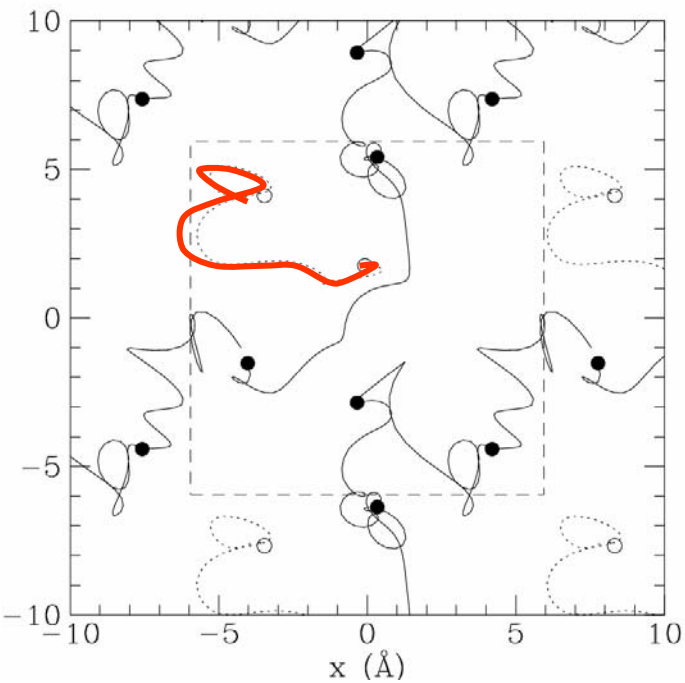
How to calculate n(r)

1. Take diagonal paths and find probability of displacing one end.

- advantage:
  - simultaneous with other averages,
  - all time slices and particle contribute.
- disadvantage: unreliable for r>Λ.

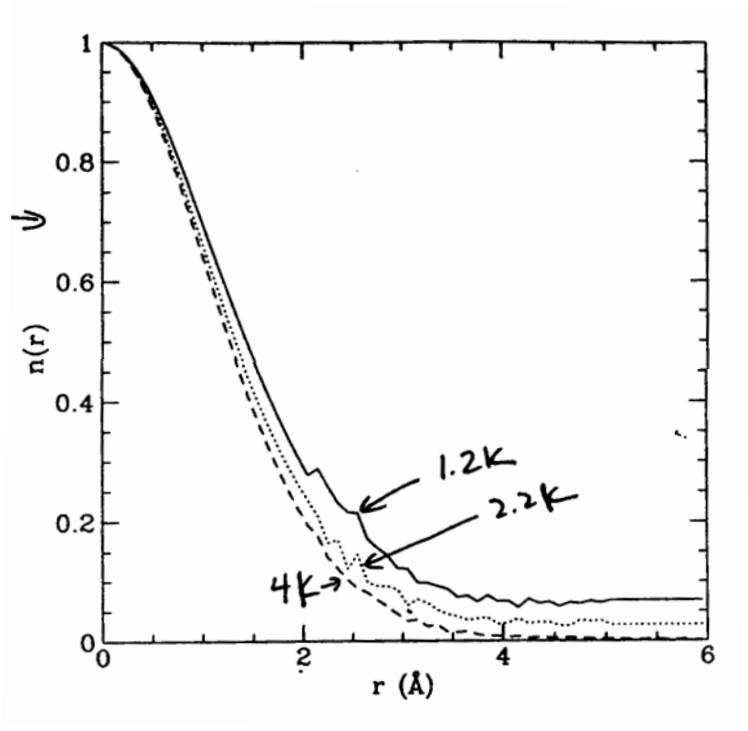
2. Do simulation off the diagonal and measure end-end distribution. Will get condensate when free end hooks onto a long exchange.

- advantage: works for any r
- Disadvantage:
  - Offdiagonal simulation not good for other properties
  - Normalization problem.

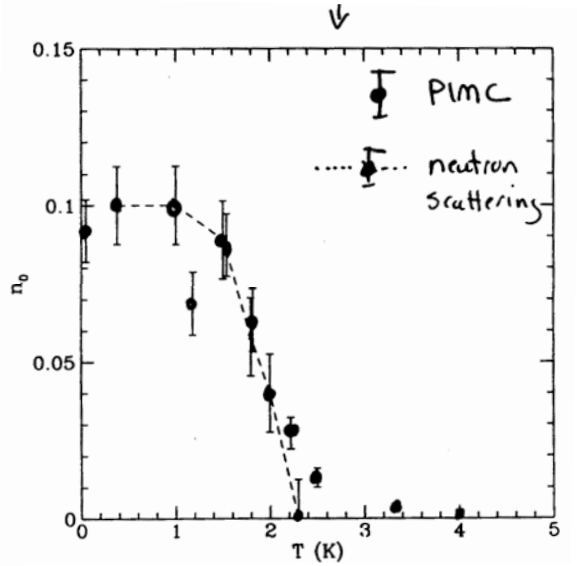


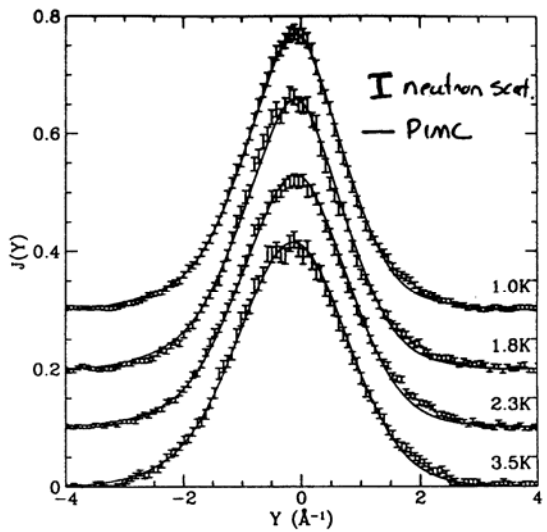
Comparison with experiment

Single particle density matrix



Condensate fraction

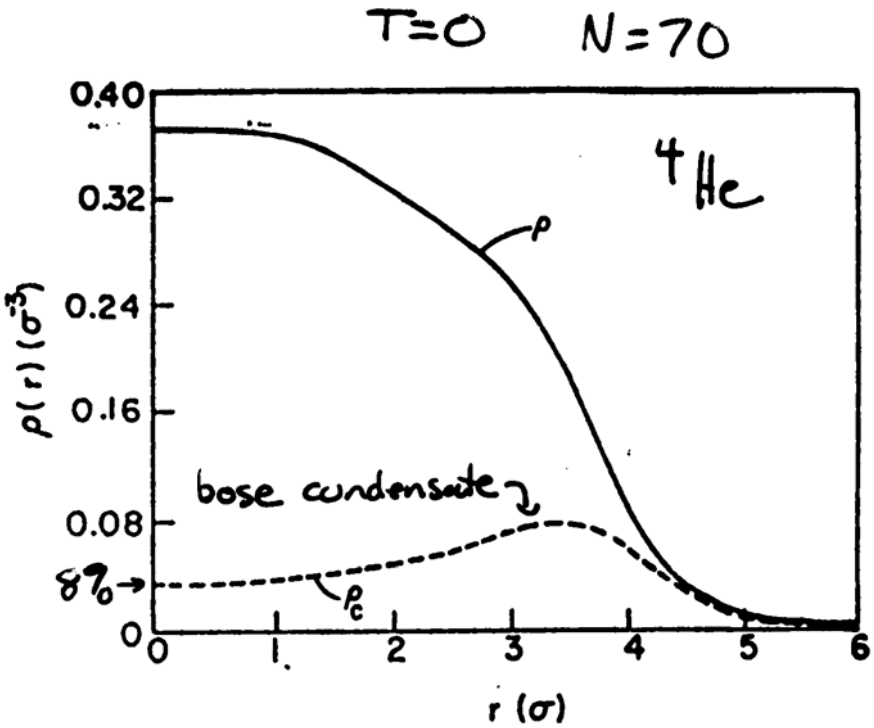




$Y = \frac{m}{\kappa}(\omega - \lambda k^2)$

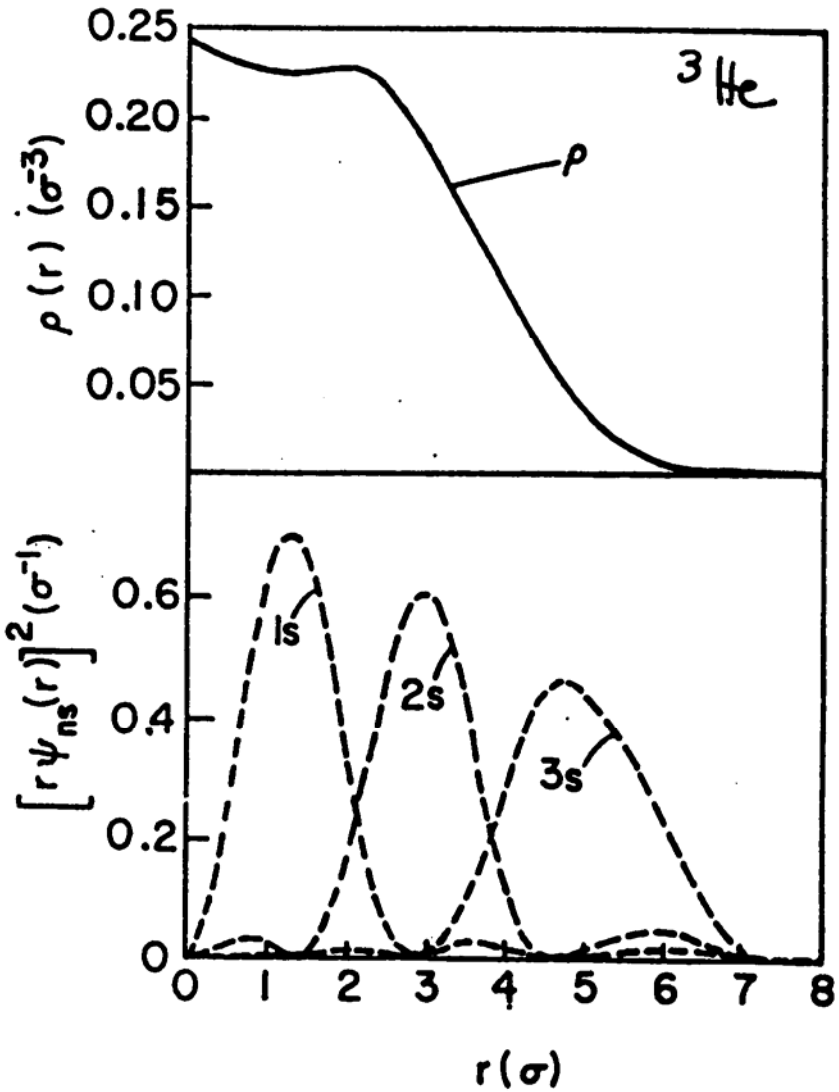
Neutron scattering cross section

Condensate in a ^{4}He droplet is enhanced at the surface



Lewart et al., PRB 37,4950 (1988).

Shell effects in ^{3}He

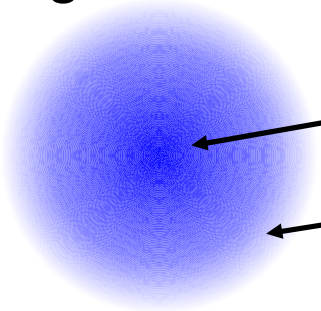


Surface of Liquid Helium

2 possible pictures of the surface

- **Dilute Bose gas model:** Griffin and Stringari, PRL 76, 259 (1996).

^{4}He



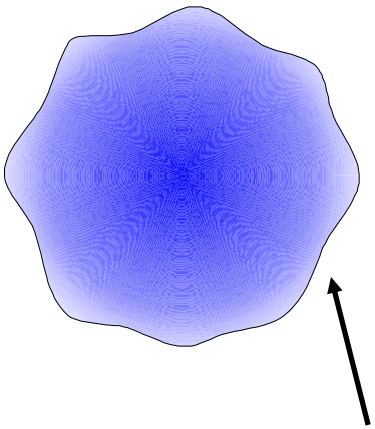
bulk: $n_0 = 9\%$

surface: $n_0 = 100\%$

- **Ripplon model:** Galli and Reatto, J. Phys. CM 12,6009 (2000).

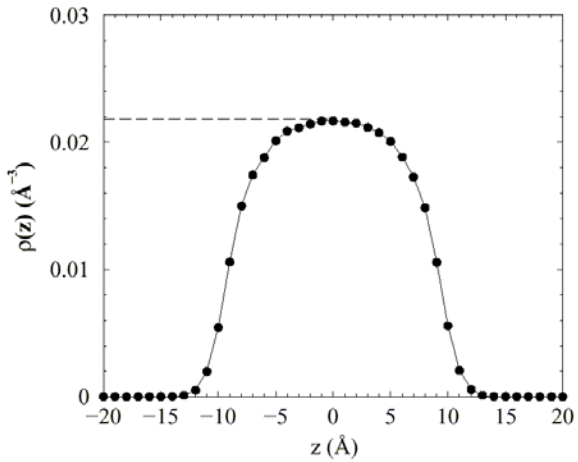
Can smeared density profile be caused by ripplons alone?

^{4}He



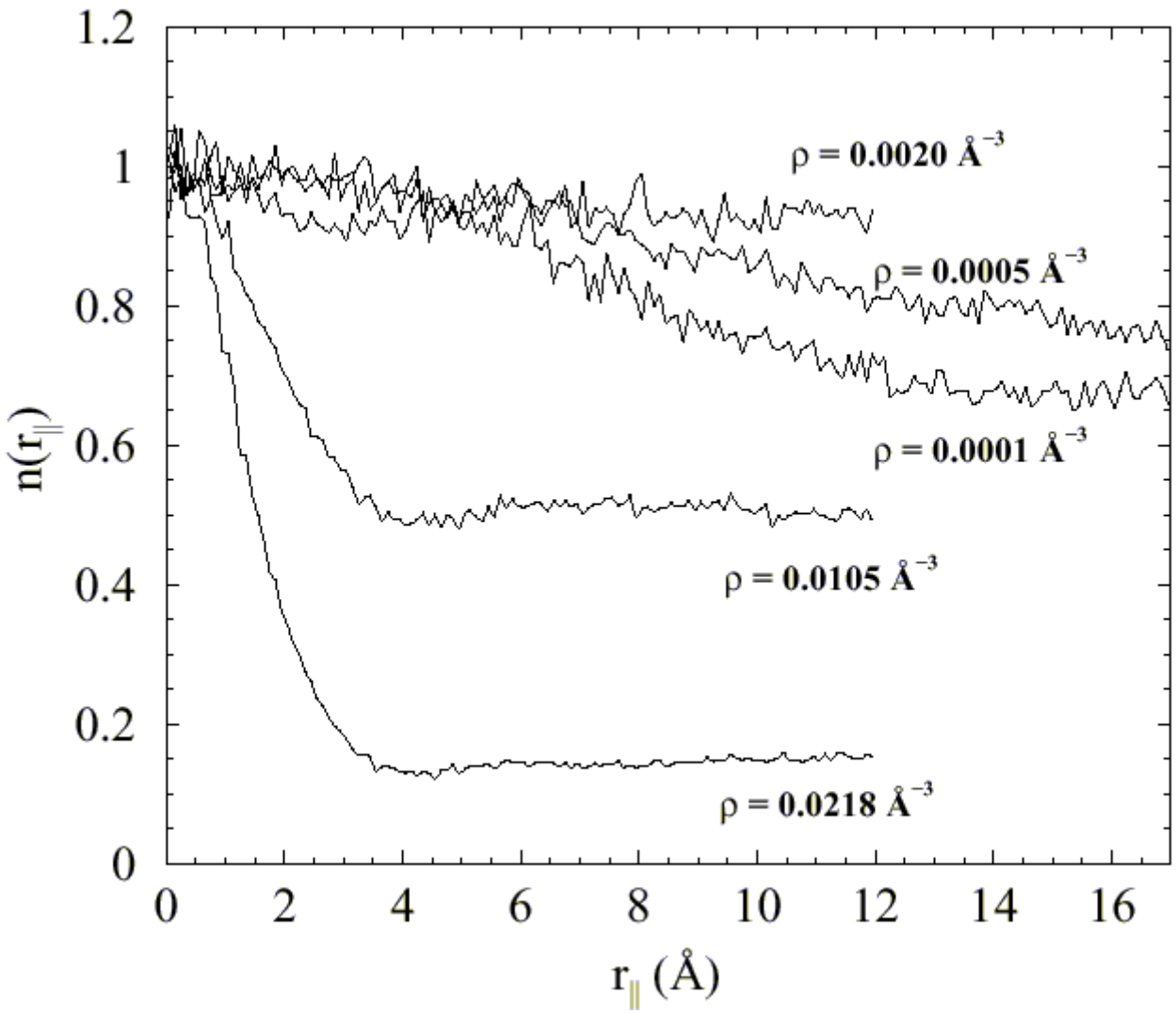
surface: $n_0 = 50\%$

Density profile does not distinguish

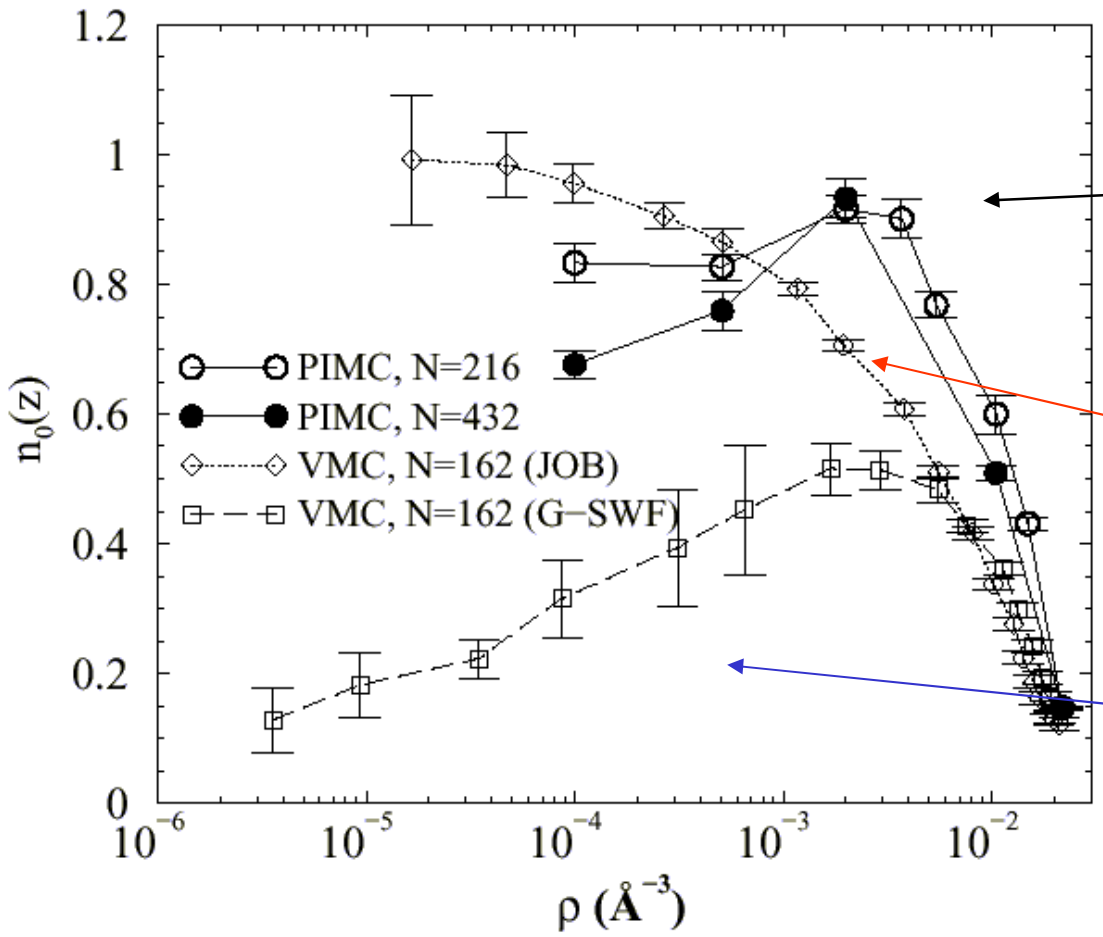


ODLRO at the Surface of Liquid ^{4}He

Simulation supports the dilute bose gas model.

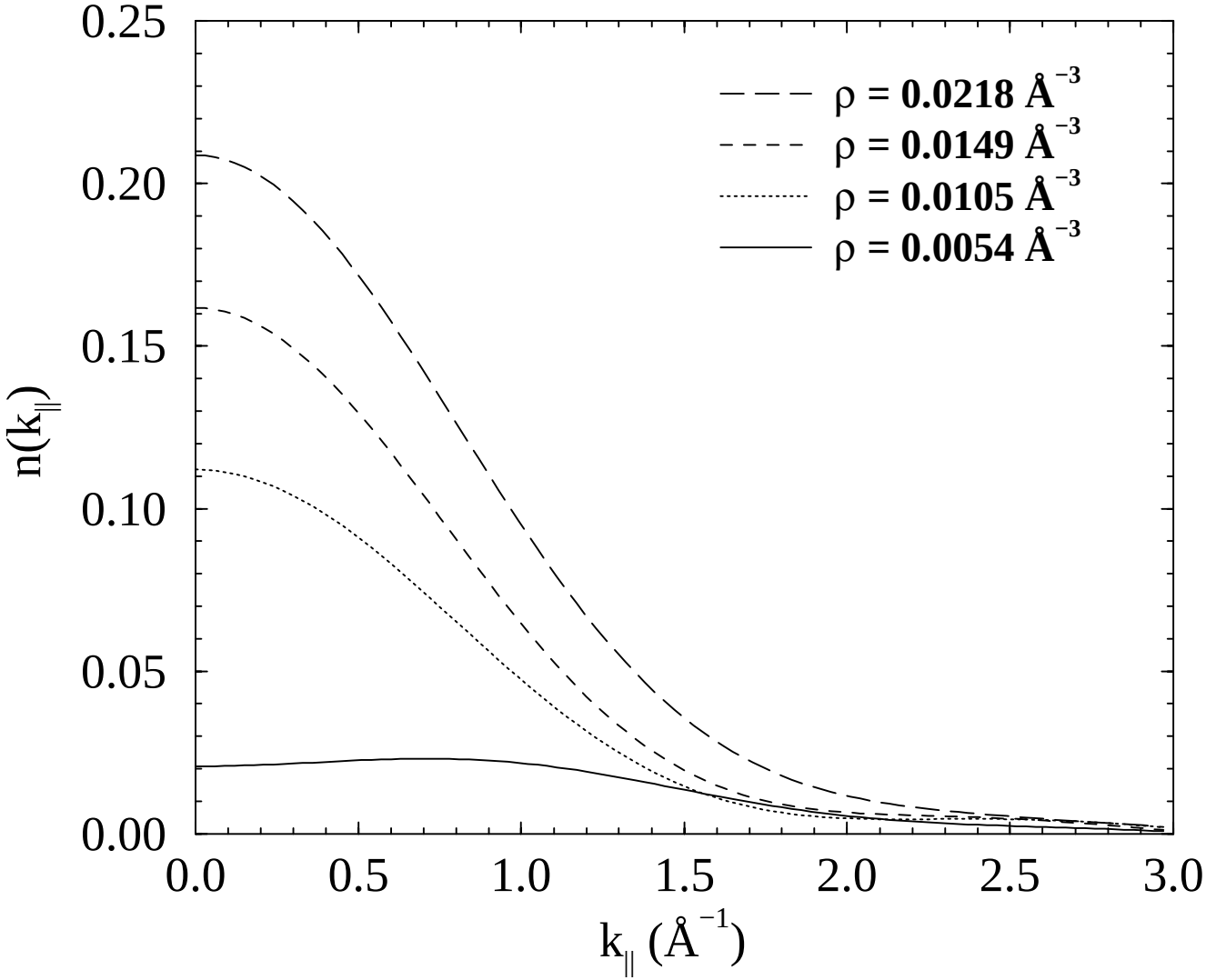


Condensate Fraction at the Surface of ^{4}He



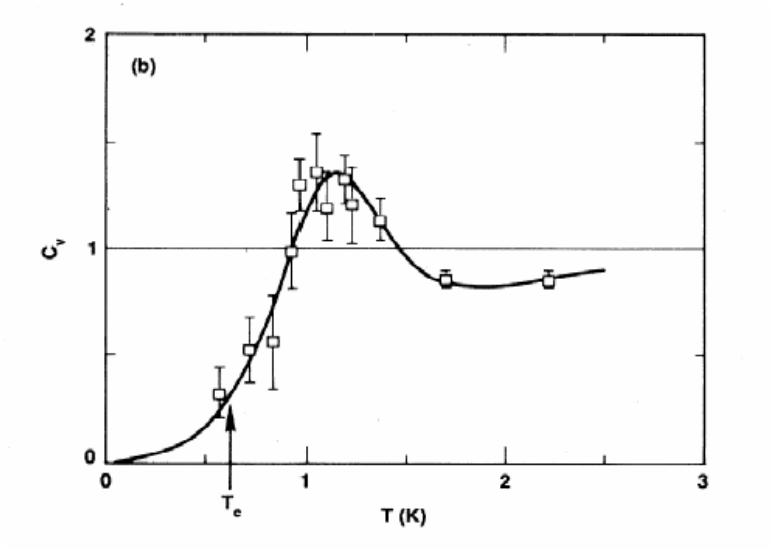
- PIMC supports dilute gas model.
- Jastrow wavefunction
- Shadow wavefunction

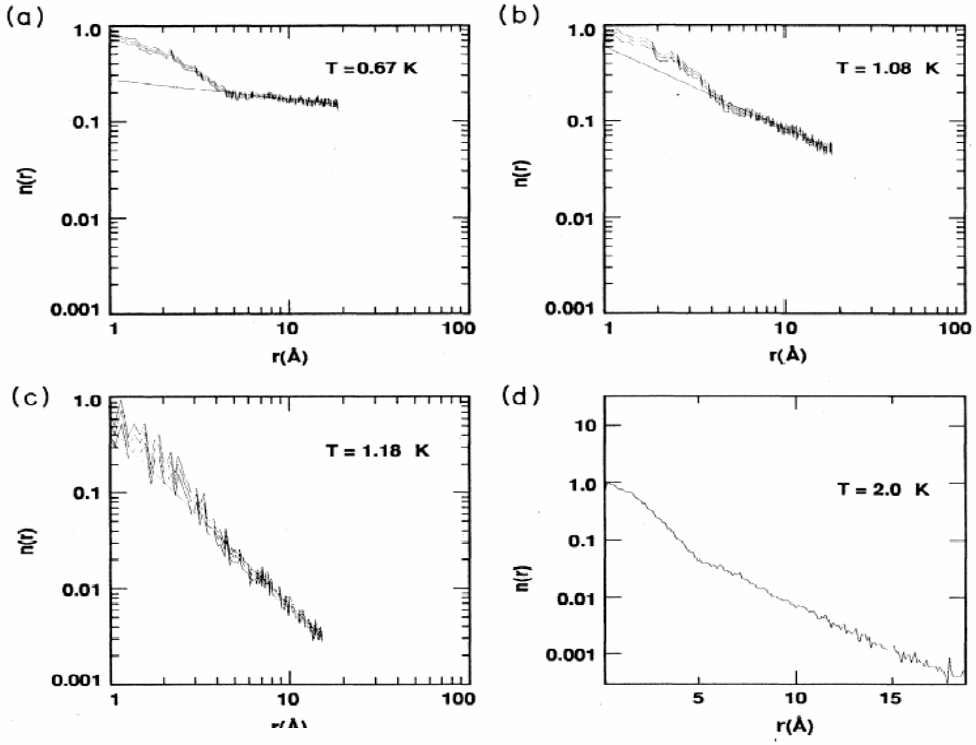
Non-condensed distribution

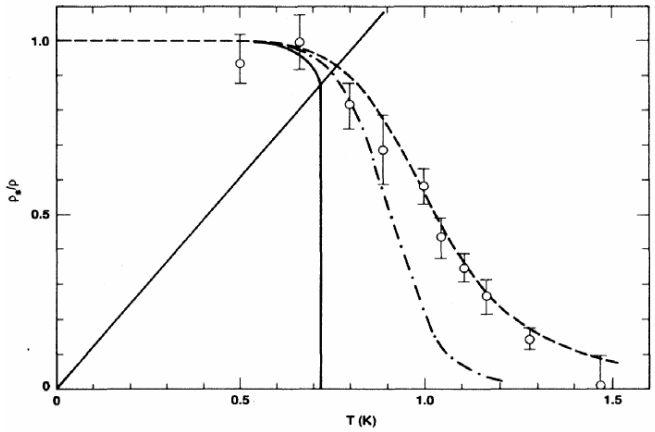


2D superfluids
Kosterlitz-Thouless transition

- Reduced dimensionality implies no bose condensate (except at T=0).
- Exchange responsible
- Specific heat bump only
- But still a good superfluid.







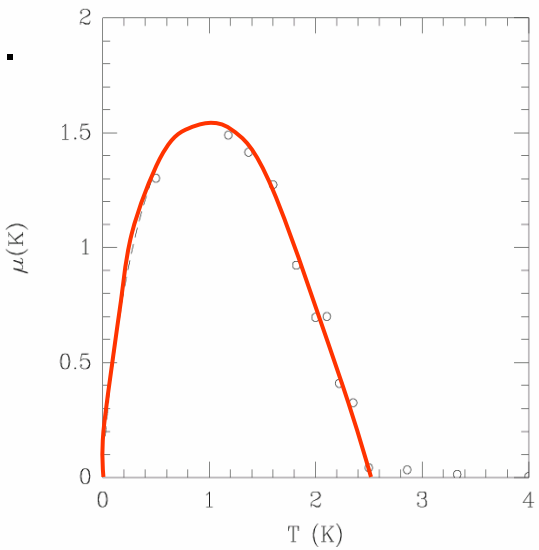
Exchange energy

- Let's calculate the chemical potential of a ^{3}He atom in superfluid ^{4}He.
- First suppose that we neglect the difference in mass but only consider effect of statistics.
- The "tagged particle" is should not permute with the other atoms.
- How does this effect the partition function?
- We do not need to do a new calculation

$$e^{-\beta\mu_3} = \frac{Z_{N-1,1}}{Z_N} = \Pr(\text{atom 1 does not exchange})$$

$$\mu_3 = \mu_4 - kT\ln\left(P_1\right) + \int_{\ln(3)}^{\ln(4)} d\ln(m)K(m)$$

- Cycle length distribution is measurable, not just a theoretical artifact.



Quantum Dynamics

- What can we do about "real-time" quantum dynamics?

$$\phi(t) = e^{-it(\mathrm{H}-\mathrm{E_T})}\phi(0)$$

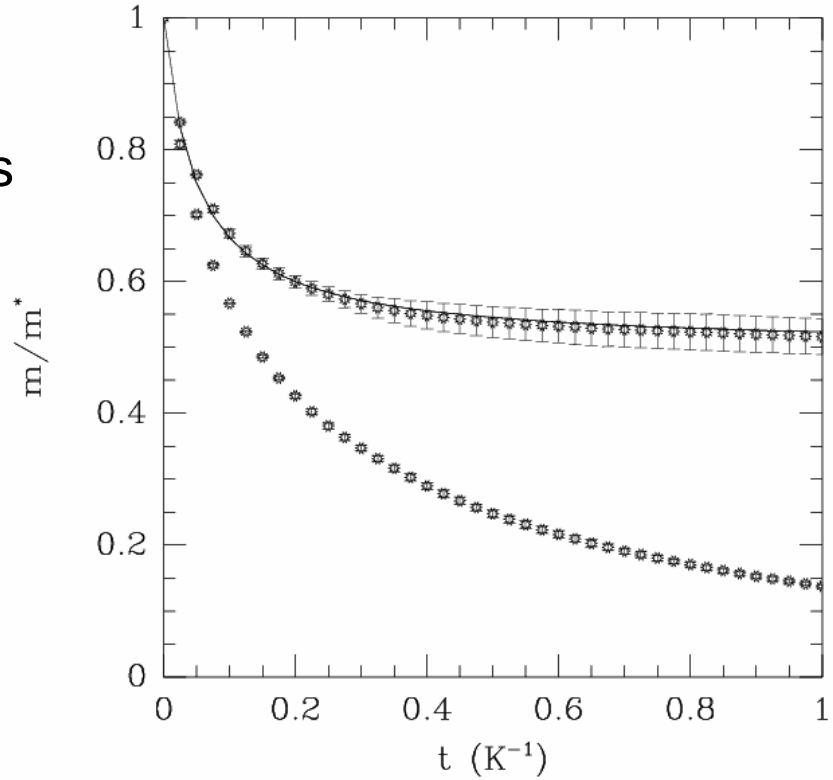
$$\phi(\tau) = e^{-\tau(\mathrm{H}-\mathrm{E_T})}\phi(0)$$

- Clearly very important!! This is what experiments usually probe.
- Feynman argued that full many-body quantum dynamics is exponentially difficult on a classical computer. Amount of memory needed to store and time needed to update, the wavefunction--which can be completely arbitrary-- grows very fast.
- Judge by the progress on quantum scattering calculations:
  - 1950-1970 2 particle problems
  - 1970-1990 3 particle problems
  - 1990-2010 4 particle problems
  - ….

Effective mass

- Effective mass is gotten from the diffusion constant at low temperature
- At short time KE dominates and $m = m^*$
- At large times, neighboring atoms block the diffusion increasing the mass by a factor of 2.
- Same formula applies to DMC!
- Lower curve is for Boltzmannons- they have to return to start position so they move less.
- Diffusion in imaginary time has something to do with excitations!

$$\frac{m}{m^*} = \frac{\left\langle \left[r(t) - r(0) \right]^2 \right\rangle}{6\lambda t}$$



Imaginary time correlations

- With PIMC (and DMC) we can calculate imaginary time dynamics: $F_O(\tau) = Z^{-1}\left\langle Oe^{-\tau\hat{H}}Oe^{-(\beta-\tau)\hat{H}}\right\rangle = \left\langle\left\langle O(t)O(t+\tau)\right\rangle\right\rangle$

  (DMC corresponds to $\beta\to\infty$)
- If we could determine this analytically we could just substitute imaginary values of τ for real values.
- Dynamic structure function is the response to a density perturbation is (e.g. density-density response)

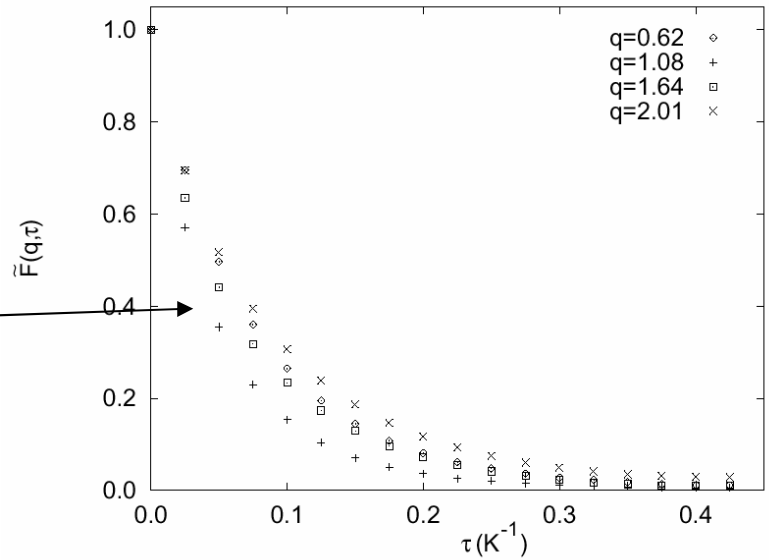
$$S_k(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt e^{i\omega t} F_k(t) \quad \text{where } O = \frac{1}{\sqrt{N}}\sum_{i=1}^{N} e^{i\vec{k}\cdot\vec{r}_i}$$

$$F_k(\tau) = \int_{-\infty}^{\infty} d\omega e^{-\tau\omega} S_k(\omega)$$

- $S_k(\omega)$ is measured by neutron scattering. We need to invert the “Laplace transform” to get $S_k(\omega)$.

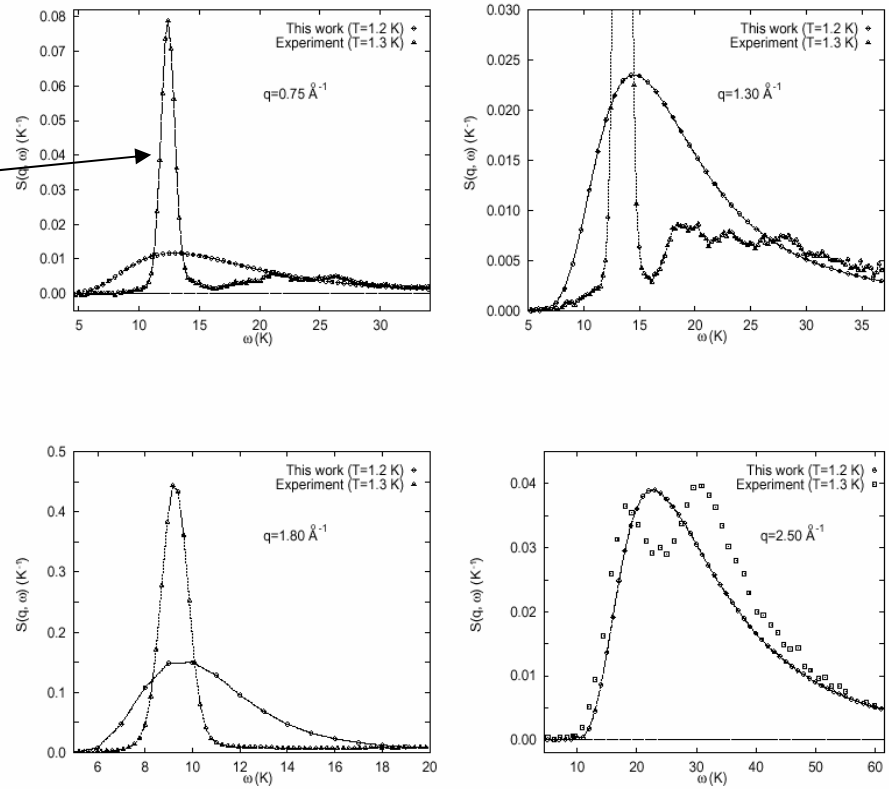
$$F_O(\tau) = Z^{-1}\left\langle Oe^{-\tau\hat{H}}Oe^{-(\beta-\tau)\hat{H}}\right\rangle$$

Little structure or information in F(q,t)



shown are different q values.

but much in S(q,w). Sharp peaks at the excitations (essentially infinitely narrow lines in the superfluid)



Bayes' theorem

- What is the most probable value of $S_k(\omega)$ given both:
  - the PIMC data, $F_k(t)$ and
  - prior knowledge of $S_k(\omega)$: e.g. $S_k(\omega) \geq 0$
- Bayes' theorem (also used by Laplace)

$$\Pr\left(S(\omega)\middle|F(t)\right) \propto P_L\left(F(t)\middle|S(\omega)\right)P_P(S(\omega))$$

- Likelihood function follows from central limit theorem:

$$P_L\left(F(t)\middle|S(\omega)\right) \propto \exp\left[-\frac{1}{2}\sum_{\tau,\tau'}\delta F(\tau)\sigma\left(\tau,\tau'\right)^{-1}\delta F(\tau')\right]$$

$$\delta F(\tau) = F\left(\tau\right) - \left\langle F\left(\tau\right)\right\rangle \text{ and } \sigma\left(\tau,\tau'\right) = \left\langle \delta F\left(\tau\right)\delta F\left(\tau'\right)\right\rangle$$

- But what to choose for the prior $P_p(S)$? Typical choice is the "entropy."

$$P_P(S(\omega)) \propto \exp\left[\alpha\sum_{\omega} S\left(\omega\right)\ln\left(S\left(\omega\right)/m\left(\omega\right)\right)\right]$$

Now two routes to making the inversion:

1. Sample $S_k(\omega)$. AvEnt Using MCMC, make moves in $S_k(\omega)$ space. Take averages and also get idea of the allowed fluctuations. Model can be defined self consistently
2. Find most probable $S_k(\omega)$. MaxEnt Maximize function. Ok if the p.d.f. is highly peaked. Estimate errors by the curvature at the maximum. Fast to do numerically but makes more assumptions.

How do we choose α? Choose it from its own prior function so the strength of the likelihood function and the prior function are balanced. Its prior function is: $P(\alpha)=1/\alpha$.

Determine MC errors by blocking and rotate to direction of independent data.

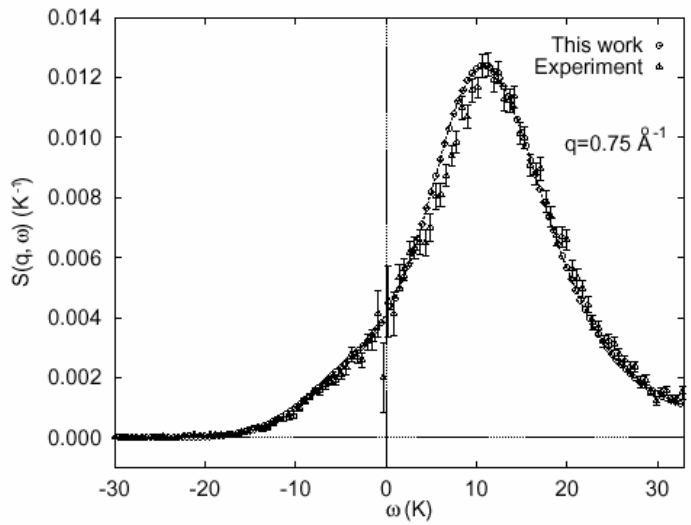
Example: Liquid ^{4}He

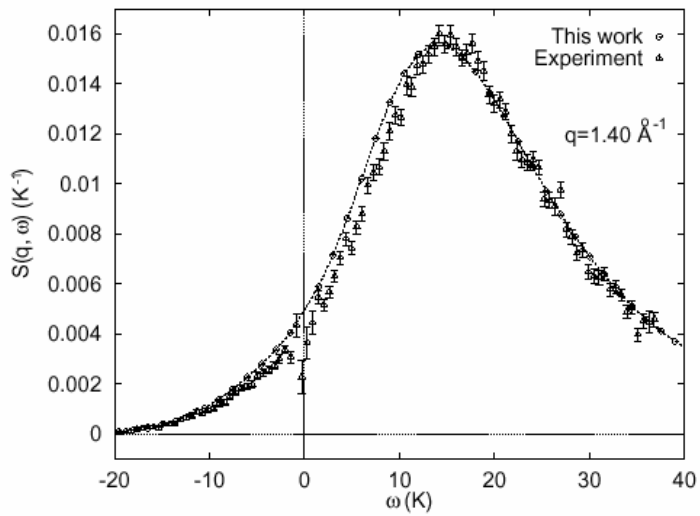
Boninsegni and DMC JLTP ***104****, 339 (1996).*

- Calculate $F_k(\tau)$ using PIMC (UPI code).
- AvEnt works beautifully in normal phase.
- Gives peaks too broad in the superfluid phase. Failure of the entropic prior.
- It makes the assumption that energy modes are uncoupled. This is false! Energy levels repel each other so that if there is energy at one level, energy levels are pushed away from nearby values.
- Would require incredible precision to get sharp features.
- But good method for determining the excitation energy.

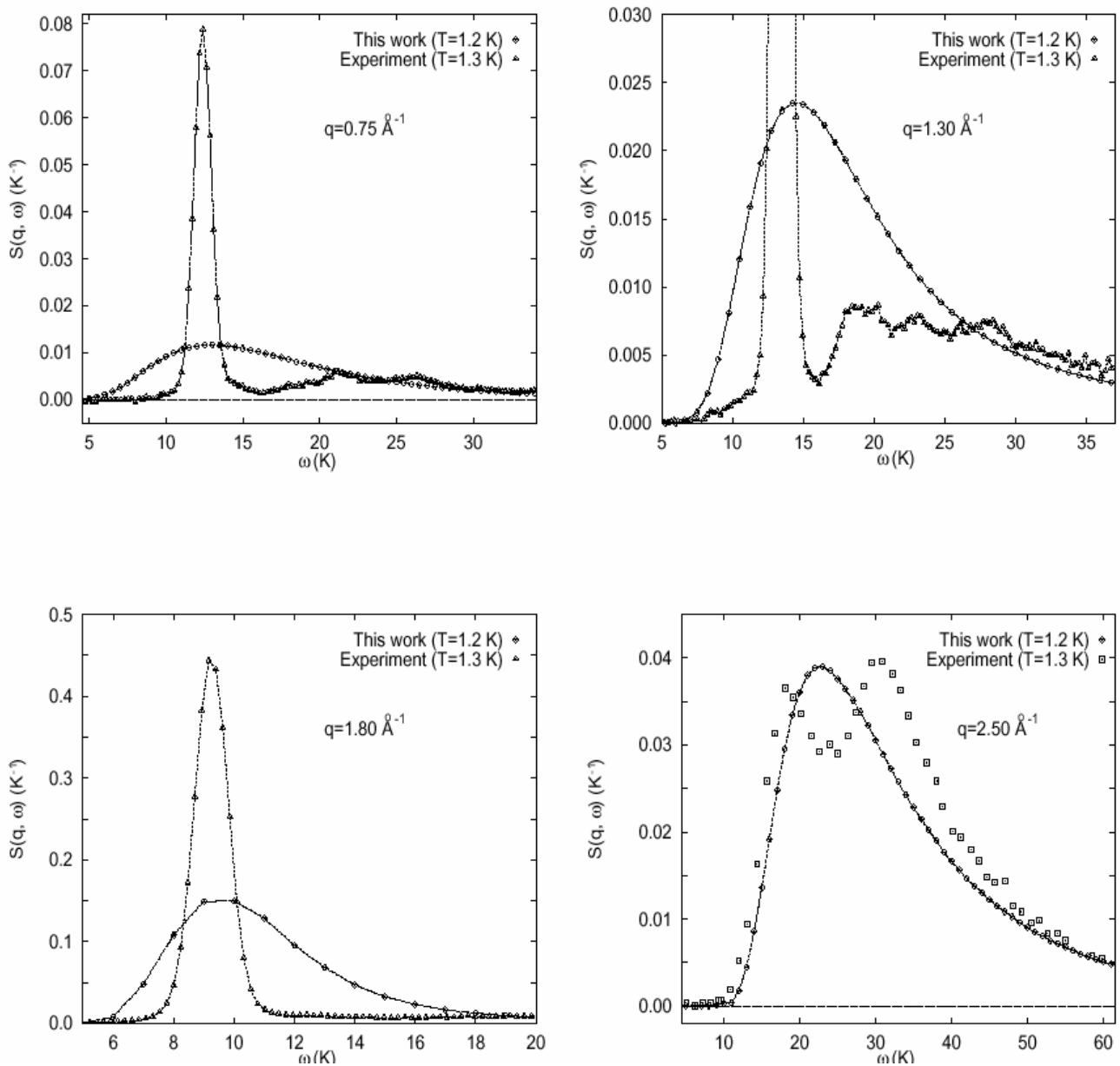
Comparison in normal liquid He phase

- MaxEnt works well in normal phase (T=4K)
- Modes are quantum but independent of each other so that max-ent prior is reasonable.





Comparison in Superfluid

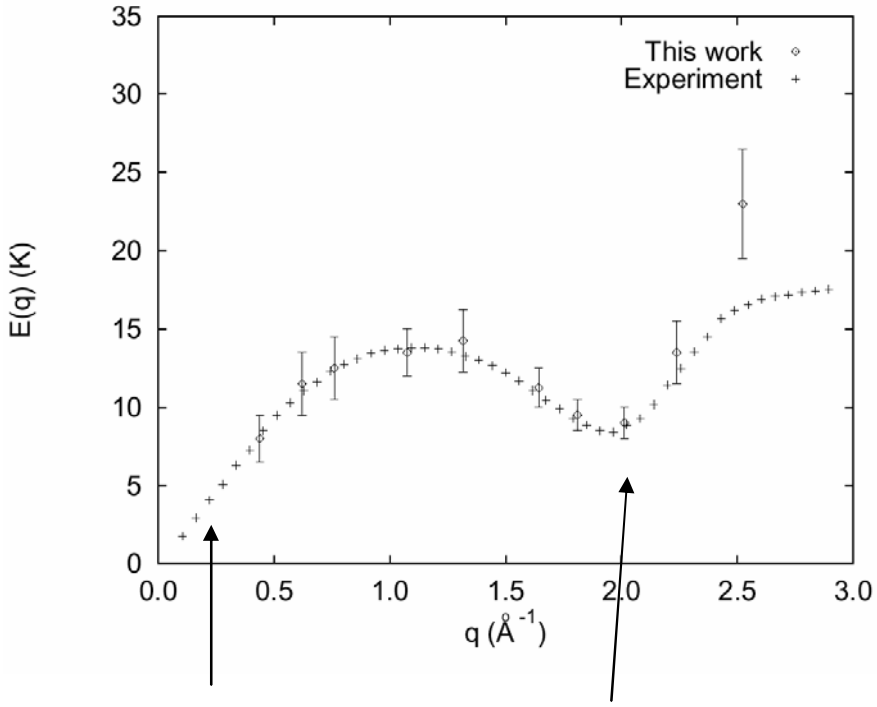


Excitation energies

Improvements?

- Better PIMC data, more time values, smaller errors.
- Work in effective hamiltonian space, not energy space.
- Get more information, for example,
  - multiphonon correlation functions
  - Incorporate exchange values
  - Analytic information about response properties

Reasonable excitation energies from MAXENT



Phonons rotons

Dictionary of the Quantum-Classical Isomorphism

Properties of a quantum system are mapped into properties of the fictitious polymer system

Attention: *some words have opposite meanings.*

Quantum	**Classical**
Bose condensation	Delocalization of ends
Boson statistics	Joining of polymers
Exchange frequency	Free energy to link polymers
Free energy	Free energy
Imaginary velocity	Bond vector
Kinetic energy	Negative spring energy
Momentum distribution	FT of end-end distribution
Particle	Ring polymer
Potential energy	Iso-time potential
Superfluid state	Macroscopic polymer
Temperature	Polymer length

Squarer produces *.dm file

Huge file contains several subfiles

- Description of grids
- Potential
- Action in various approximations
- Action at various time slices
- Beta or mass derivatives of the action
- Sampling tables

- Radial grid has several options. Can vary for each type of pair interaction to put more grid points in some region.
  - GRID n LINEAR r1 r2
  - GRID n LOG r1 r2
  - .....

Density matrices used in PUPI

- Pair density matrix: generated by squarer, (see new simpler code on WEB) Includes expansion for offdiagonal components. Action is evaluated in **dmeval** (calls **dstnce** and **offd**). Energy is evaluated in **pairact**. Calls radial tables for variety of grid types.
- Single particle propagator in periodic boundary conditions: **fpdmg.f** uses a table set up in **setfpdm.f** Used for fermion and boson density matrix. Also to determine permutations.
- Free fermion density matrix. **Ofill** sets up the matrix, calls **invert or newup** to calculate the determinant.
- Particle near a plane (image method). **Dleak.f** (related computations in the nodal action, **dnode.f**)
- Long range potential: (in k-space) (**ewald.f**) setup code generates the expansions. Uses fourier coefficents (rhok).

Functionality:

- Action at specified temperatures differing by powers of 2 in temperature
- beta derivative
- spatial derivative
- has to be flexible with respect to grid type, analytic features ...
- update formulas for all cases—how we compute changes with respect to a single variable and derivatives.