beam to achieve steady state in our apparatus. This is in agreement with a theoretical estimate of $\langle \Delta v^2 \rangle / \langle v \rangle^2$, using Eqs. (9) and (13) and the experimental parameters.

[7] The adiabatic law for the cloud of vortex rings can be obtained in a way similar to the derivation for an ordinary gas. If we multiply Eq. (1) by pv and integrate over p using $\Delta p \sim -2(\Delta v)v^{-3}$ and the symmetry of the distribution of Δp and other conservation forms, Eqs. (3) and (6), then the relation $P \sim n^{3/2}$ is easily obtained.

Quasiclassical and Quantum Degenerate Helium Monolayers*

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He^3 and He^4 adsorbed on graphite have heat capacities qualitatively different from monolayers on other substrates. Both isotopes have quasiclassical two-dimensional gas behavior at low fractional coverage x and $T \gtrsim 2$°K. At lower T and $0.14 \leq x \leq 0.39$, He^3 shows quantum degeneracy, with Fermi temperatures of several degrees Kelvin. The specific heat per atom of He^4 at the same coverages rises above k and has pronounced peaks at low T.

Helium monolayers and fractional monolayers have low-temperature heat capacities whose characteristics have led to the belief that the films form two-dimensional solids. Since films adsorbed on different substrates[1-6] have qualitatively similar characteristics at comparable coverages, it has seemed that the thermodynamic properties of the monolayers are primarily determined by the He, the substrate being mainly an attractive surface for adsorption. However, difficulties arise if the substrate is relegated to so passive a role. He^4 on Ar-plated Cu has two-dimensional (2D) solidlike heat capacities at coverages as low as 0.1 monolayer,[4] implying a latent heat of sublimation from 2D solid to 2D vapor much larger than estimated for direct He-He interactions[7,8] or to substrate-phonon-enhanced interactions.[9] A further puzzle is the absence of thermal evidence for a 2D liquid phase,[10] which appears to be the preferred form of the condensed state.[8] To explain the large apparent 2D binding, Roy and Halsey[11] suggest that substrate inhomogeneities tend to force adatoms into dense surface aggregates: The heat capacities of such aggregates might then correspond to 2D solids under high lateral "pressures."

If previous studies have been strongly influenced by substrate inhomogeneity, He adsorbed on graphite might yield markedly different characteristics. Ar and Kr on graphitized carbon black[12] show stepwise vapor-pressure isotherms instead of the usual Brunauer, Emmett, and Teller type, indicating a high degree of surface uniformity. Vapor pressures are less sensitive to substrate variations than is heat capacity, but the stepwise isotherms offer the possibility that graphite approximates a uniform substrate for heat-capacity studies. We have undertaken such a study, and our results for He^3 and He^4 are spectacularly different from previous work on other substrates.

This Letter reports measurements at low to moderate fractional monolayer coverages and temperatures from 1.1 to 4.2°K. Details of calorimeter construction will be given elsewhere, but we give here the essential characteristics of the adsorption cell. The graphite is in the form of exfoliated pyrolytic foils[13] packed into a thin-walled Cu cylinder of 12.6-cm^3 volume. The cell was baked at 900°C in vacuum and subsequently protected under inert atmospheres. We obtained a 77°K Ar isotherm having similar shape to those on graphitized carbon black. The first step is at 70 cm^3 STP; using 12.8 Å^2 as the molecular area of Ar,[14] the effective adsorption area is $A = 240$ m^2. This area is considerably greater than the areas of Cu sponge in cells of the same volume.[3,4] A He^3 isotherm at 4.2°K shows a step at 90 cm^3 STP which we attribute to the completion of its first layer. Thermal relaxation times are comparable to the relaxation times for Cu cells.

The heat capacity of the "empty" cell is shown in Fig. 1. Values agree with estimates based on its content of graphite and Cu. Addition of relatively small amounts of either He^3 or He^4 increases the signal substantially. In Fig. 1 we also present the total heat capacity of the cell containing fractional coverages $x = 0.15$ of He^4 and $x = 0.14$ of He^3 (where x is the ratio of adsorbate quantity to He^3 monolayer capacity). Assuming that the heat capacity of the film is equal to the

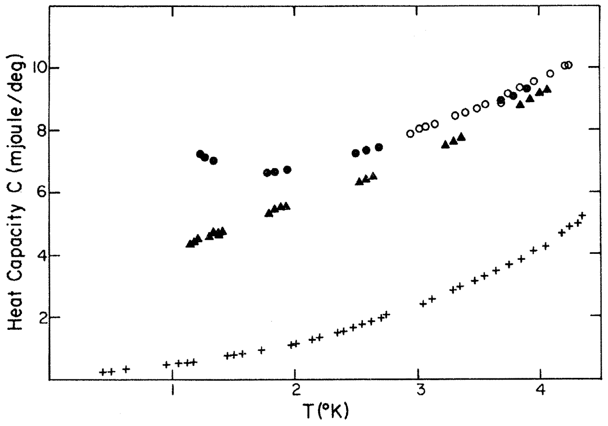

FIG. 1. Total heat capacities: plus signs, empty calorimeter; open circles, 0.148 He^4; closed circles, 0.153 He^4; closed triangles, 0.143 He^3.

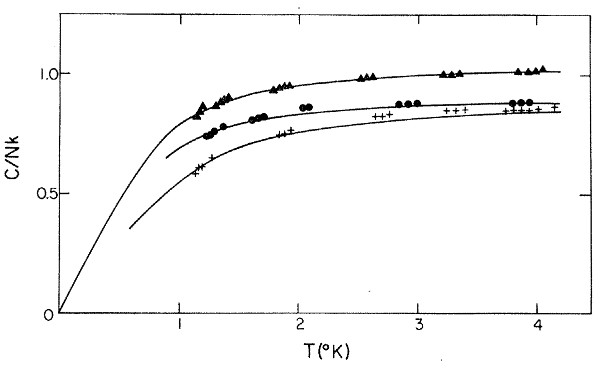

FIG. 2. Specific heats of He^3 films: closed triangles, $x=0.14$; closed circles, $x=0.26$; plus signs, $x=0.39$. Solid lines are theoretical curves for ideal, 2D Fermi gases with parameters listed in Table I.

difference between total and "empty" signals, we find that the two isotopes have the same specific heats per atom at higher temperatures: 1.03k for He^4 and 1.02k for He^3 at 4°K. The specific heats diverge gradually at decreasing T, He^4 rising to a specific heat per atom of about 1.3k at 1.2°K and He^3 falling below k by a comparable factor. These trends are amplified at higher x, where we find that He^4 develops pronounced peaks between 1 and 2°K. In Fig. 2 we present results for He^3 up to $x=0.39$, and in Fig. 3 He^4 data at the same coverages.

We believe that the results lead to four main conclusions, outlined below.

(a) The low-x, high-T results closely approximate the constant specific heat per atom k characteristic of low-density, 2D gases. Such behavior implies that the surface mobility of the He is so rapid that the lowest translational bands have either little or no energy gap between them, for otherwise the heat capacity would have broad maxima and minima at values of kT of the order of the bandwidths and interband gaps.[15] Calculations for He^4 on (100) faces of solid rare gases[16-18] show well-separated first and second bands, but for He^4 on Ar-plated Cu and Xe-plated graphite[19] the lowest bands partially overlap, and a corresponding calculation for He on graphite is almost certain to show considerable overlap.

(b) Long-range variations in adatom-substrate interaction energies are small relative to kT. This is based on a calculation for classical noninteracting adatoms on a smooth plane having a gradual variation of adsorption energy along the surface. The variation produces a Schottky-like high-temperature tail behaving as $(\Delta\epsilon_0/kT)^2$ for total inhomogeneity $\Delta\epsilon_0 \ll kT$. We examined both He^3 and He^4 in the quasiclassical regime above 2°: The absence of a definite T^{-2} signal indicates that $\Delta\epsilon_0/k \lesssim 5$°K.

(c) There is no unusual enhancement of He-He interactions at the experimental densities and temperatures. This is deduced from the lack of evidence for condensation, which would give a discontinuous rise in heat capacity and dependence as exp($-$const/T) at lower T. The peaks in He^4 do not have such shape and the He^3 data fall monotonically with T. Theoretical estimates of ground-state energies of 2D He^3 and He^4 give comparable binding per atom of ~0.6°×k,[7,8] implying that both isotopes would condense at roughly the same conditions of temperature (<1°K) and density.

(d) Statistical interactions are unimportant at low x and high T, but are responsible for the differences between He^3 and He^4 at low T. This conclusion is consistent with the magnitude of the

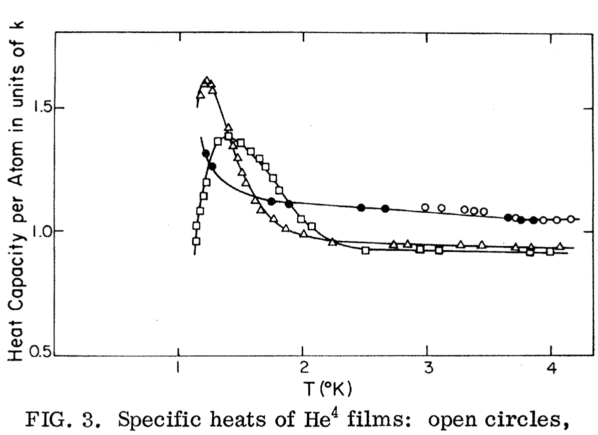

FIG. 3. Specific heats of He^4 films: open circles, $x=0.148$; closed circles, $x=0.153$; open triangles, $x=0.255$; open squares, $x=0.372$.

degeneracy parameter $\rho\lambda^2$ [ρ is the areal density N/A and λ is the thermal wavelength $h(2\pi mkT)^{-1/2}$]. Assuming m to be the free-atom mass, we calculate $\rho\lambda^2 \simeq 0.3$ for He^4 at $x = 0.15$, $T = 4°$, while $\rho\lambda^2 \simeq 1$ for He^4 at $x = 0.39$, $T = 2°$. Since the transition between quasiclassical and quantum-degenerate regimes should occur at $\rho\lambda^2 \sim 1$, the magnitudes of the degeneracy parameter are consistent with our interpretation.

We therefore explored the 2D-gas model further, testing the correspondence between experimental results and the properties of 2D ideal quantum gases. For He^3 the model is a natural one, for it is well known that the heat capacity of a 2D fermion gas falls monotonically as the degeneracy parameter increases. We find that the theory[15,20] can provide a good overall fit to each of the samples, as shown in Fig. 2. The theoretical curves were computed by treating particle number N and Fermi temperature $T_\mathrm{F}(0)$ as adjustable parameters for each sample. The adjusted N's are in moderate agreement with the actual quantities, but the Fermi temperatures are significantly higher than given by ideal-2D-gas theory. Higher $T_\mathrm{F}(0)$ implies effective mass $m^* < m$, adatom interactions, or lateral fields (see following Letter[21]). The second and third mechanisms reduce the effective area per atom. Parameters for each sample are listed in Table I.

The simple 2D-gas model is qualitatively unsuccessful for low-T He^4: It predicts the same T dependence for bosons as for fermions, with magnitudes differing only by mass and nuclear-spin factors.[15,20] Yet we find that when He^3 begins to fall appreciably below the specific heat per atom k, He^4 has a comparable rise. It therefore seems that although known 2D theories fail, statistical effects must play as important a role for He^4 as they appear to for He^3. A simple but essential modification of the standard theory, offering a qualitative explanation of He^4, is described in the accompanying Letter.[21]

We are extending the experimental study to higher coverages than those reported here, and we find novel effects not suspected from the low-x results. These findings will be published soon.

We are grateful to Professor D. B. Fischbach for instructive conversations concerning graphite and for suggesting the "Grafoil" material used in this experiment.

Table I. Parameters of He^3 samples.

Measured quantities		Fitted parameters	
x	N (10^{20})	$T_\mathrm{F}(0)$ (°K)	N (10^{20})
0.14	3.47	3.6	3.33
0.26	6.17	3.4	6.85
0.39	9.37	4.8	10.6

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