Effects of Lateral Substrate Fields on Helium Monolayers*

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Heat capacities of He monolayers on graphite show qualitative differences between He^3 and He^4, contrary to the usual theories of two-dimensional quantum gases. We attribute the differences to weak lateral fields due to substrate inhomogeneities, and using a two-dimensional ideal-gas model show qualitative agreement between theoretical and observed heat capacities.

Pronounced peaks in the heat capacities of submonolayer He^4 films on graphite have been observed by Bretz and Dash.[1] The heat capacities of similar He^3 films do not display similar behavior. Such results immediately invite speculation on the occurrence of a Bose condensation in the He^4 film. This possibility has received much attention in the last few years. It has been well established that Bose condensation, defined by the macroscopic mean occupation of a single-particle state, can be excluded at finite temperature in an infinite two-dimensional system in which the density is everywhere bounded.[2,3] It is likewise well established that such a condensation cannot be excluded in a finite two-dimensional system.[4] In this Letter we present arguments against the relevance of finite geometrical effects in the recent experiments, and we propose an alternative mechanism for the observed behavior.

A uniform, two-dimensional, ideal Bose gas of finite area A exhibits a condensation as defined above at a temperature which is given approximately by

$$T_0 \approx 2\pi\hbar^2\bar{n}/km \ln N,$$

where $\bar{n} = N/A$ is the areal density. Assuming that the order of magnitude of the transition temperature in a He^4 monolayer can be estimated by the above expression, one finds with $A = 240$ m^{-2} and $\bar{n} = 0.026$ Å^{-2} (parameters appropriate to the $x = 0.255$ monolayer coverage film of Ref. 1) that $T_0 \approx 0.02$°K. This is two orders of magnitude smaller than the temperatures corresponding to the observed peaks in the He^4 heat capacity. Alternatively, if we assume that the film is actually composed of much smaller regions (presumably because of cracks and major imperfections of the substrate), we find that the characteristic dimensions of the individual regions must be 10 Å in order to produce T_0's comparable to the observed peak temperatures. However, x-ray diffraction studies of the substrate material yield linewidths corresponding to crystallite dimensions greater than 400 Å and optical examination indicates that the graphite planes are ordered over as much as 10^4 Å. It appears unlikely, therefore, that the specific heat is signaling a condensation due to the finite size of the sample.

Another possible explanation of the experimental results, which is explored more fully below, can be summarized as follows. Widom[5] has shown that an ideal, two-dimensional Bose gas, in the presence of external fields, can exhibit a condensation at finite temperatures even in the thermodynamic limit. The density, however, becomes unbounded somewhere in the system at the critical temperature.[3] Presumably, interparticle interactions, which will serve to restore a finite density everywhere, will cause the transition and associated singularities in the heat capacity to vanish. However, it is to be expected that an increase of heat capacity with decreasing temperature will remain as a remnant of the ideal transition. Further, the data of Ref. 1 indicate that the submonolayer systems behave like ideal gases at high temperatures. Thus one expects the heat capacity of the interacting system to be well approximated by that of the ideal system down to temperatures in the immediate vicinity of the ideal critical temperature where the increase in density somewhere in the system makes it necessary to include effects of interparticle interactions.

As to the origin of the external fields themselves, Widom considered the cases of rotation and gravitation. In the monolayer experiments, these field sources may be completely neglected. However, we consider a third possibility which appears to be present in virtually all experimental films. These are the lateral fields arising from substrate inhomogeneities, which cause the adatom-substrate binding to vary from point to point along the surface. Indeed, the qualitative differences between the present experiments and previous studies is attributed to the strong inhomogeneities of substrates previously used and the closer approach of the graphite surfaces to the uniform ideal. But it is reasonable to expect that the graphite has some residual inhomogeneity, and in the following we outline a calculation indicating that even quite small fields have a profound effect on the Bose system.

Consider a two-dimensional ideal gas adsorbed on a substrate of area πR^2 and subject to a macroscopic inhomogeneity in the substrate potential. The properties of such a system may be obtained from the quasiclassical approximation to the distribution in phase space:

$$dn_\pm(\vec{r},\vec{p}) = \frac{d^3p\,d^3r}{2(2\pi\hbar)^2}\,\frac{(3\pm 1)}{\exp\beta[H(\vec{p},\vec{r})-\mu]\pm 1}, \qquad (1)$$

where the plus (minus) sign is taken for Fermi (Bose) statistics and $H(\vec{p},\vec{r})$ is the classical Hamiltonian of the single particle. The above approximation is derived for the Bose system by writing the logarithm of the grand partition function as

$$\ln Z = -\sum_i \ln\{1-\exp[-\beta(E_i-\mu)]\},$$

where the E_i are the eigenvalues of a single particle in the potential V due to the inhomogeneity. Following Widom, we expand the above as $\ln Z = \sum_1^\infty n^{-1}e^{n\beta\mu}Q(n\beta)$, where $Q(n\beta)=\sum_i \exp(-\beta E_i)$ is the single-particle partition function in the canonical ensemble evaluated at an effective temperature T/n, T being the temperature of the Bose system. This quantity can be expanded as a power series in $\hbar$, with the leading term being the single-particle classical partition function.[6] The higher-order terms involve classical averages of derivatives of the single-particle potential and may be neglected as V is taken to vary over macroscopic distances. Substituting the classical single-particle partition function into the expansion of $\ln Z$ and interchanging orders of summation and integration yields

$$\ln Z = -\iint \frac{d^3p\,d^3q}{(2\pi\hbar)^2}\ln\{1-\exp\beta[\mu-H(\vec{p},\vec{q})]\}. \qquad (2)$$

The quasiclassical distribution of Eq. (1) for the Bose system immediately follows from the appropriate derivative of the above result. The derivation of the Fermi result makes use of the same expansion in $\hbar$ although the grand partition function itself rather than its logarithm is used to generate an expression in terms of Q. The result is the same as Eq. (2) except for a spin factor of 2 and a plus sign preceding the exponential

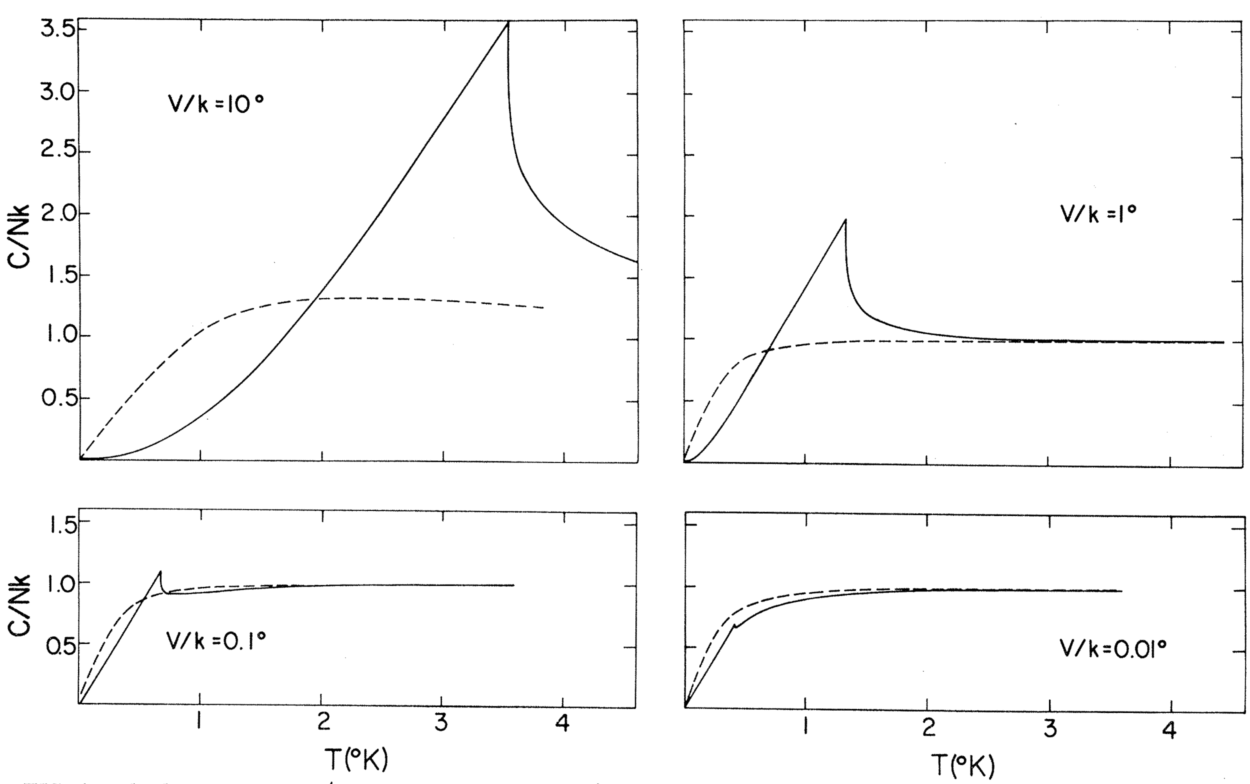

FIG. 1. The behavior of C_+/Nk (dashed line) and C_-/Nk (solid line) for four values of $V(R)$. The average density is 0.026 $\mathrm{\AA}^{-2}$.

and integrals. For definiteness we henceforth take the potential due to the inhomogeneity to be harmonic: $V(r)=\frac{1}{2}Kr^2$, $r^2=x^2+y^2$. In this case the density $n_\pm(r)$, average density $\bar{n}_\pm$, and heat capacity at constant area $C_\pm$ are[7]

$$n_\pm(r)=\pm(3\pm1)(2\lambda^2)^{-1}\ln\{1\pm\exp[-\beta(\tfrac{1}{2}Kr^2-\mu)]\}, \tag{3}$$

$$\bar{n}_\pm=(3\pm1)(2\lambda^2\xi)^{-1}[F_{2\pm}(\alpha)-F_{2\pm}(\alpha+\xi)], \tag{4}$$

$$\frac{C_\pm}{Nk}=\frac{(3\pm1)}{2\lambda^2\bar{n}\xi}\left\{[6F_{3\pm}(\alpha)-6F_{3\pm}(\alpha+\xi)-4\xi F_{2\pm}(\alpha+\xi)-\xi^2F_{1\pm}(\alpha+\xi)]-\frac{[2F_{2\pm}(\alpha)-2F_{2\pm}(\alpha+\xi)-\xi F_{1\pm}(\alpha+\xi)]^2}{F_{1\pm}(\alpha)-F_{1\pm}(\alpha+\xi)}\right\}.$$

In the above equations, λ is the thermal wavelength $h(2\pi mkT)^{-1/2}$, $\xi=\beta V(R)$, $\alpha=-\beta\mu$, and $F_{\sigma\pm}$ are the usual Bose and Fermi functions defined by

$$F_{\sigma\pm}(\alpha)=\Gamma^{-1}(\sigma)\int_0^\infty dy\, y^{\sigma-1}[\exp(y+\alpha)\pm1]^{-1}.$$

The Bose gas condenses below a critical temperature T_c given implicitly by the relation

$$\bar{n}=(\lambda_c^2\xi_c)^{-1}[F_{2-}(0)-F_{2-}(\xi_c)].$$

For $\xi_c\ll1$, the critical temperature is given by

$$T_c\approx(2\pi\hbar^2\bar{n}/km)\{1+\ln[2\pi\hbar^2\bar{n}/mV(R)]\}^{-1}.$$

Below T_c it is understood that the expressions for $n_-(r)$, $\bar{n}_-$ of Eqs. (3) and (4), refer to the uncondensed phase. As T_c is approached from above, the derivative of the specific heat C_- diverges and the density at the origin becomes unbounded, as is seen from Eq. (3) with μ set equal to zero. At high temperatures both heat capacities behave as $C_\pm\to1+V^2(R)/12(kT)^2$. As C_+ vanishes linearly with T, the inhomogeneity manifests itself in the Fermi system by a maximum in the specific heat.

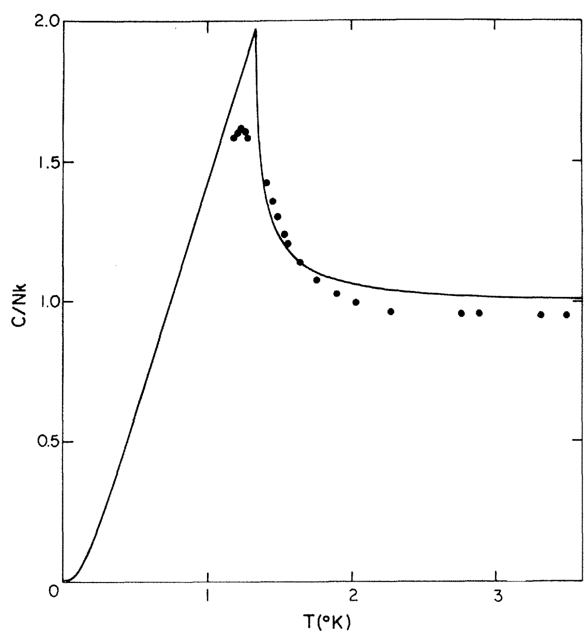

FIG. 2 Experimental data compared with C_-/Nk with $V(R)/k=1°$ and average density of 0.026 Å^{-2}.

In Fig. 1 we show C_+/Nk and C_-/Nk for several values of $V(R)$ and with $\bar{n}=0.026$ Å^{-2} (which, again, corresponds to the fractional monolayer coverage $x=0.255$ of Ref. 1). In Fig. 2 experimental data from Ref. 1 are compared with C_-/Nk evaluated with the same average density and $V(R)/k=1°$. For purposes of orientation, at $T=1.5°$, the density at the origin, as obtained from Eq. (3), is $n_-(0)=0.06$ Å^2 which corresponds to an interparticle separation which is more than 1.5 times as large as the helium hard-core diameter. The density $n_+(0)$ does not attain the above value at any temperature with the above parameters.

Clearly one could obtain a better fit to the data by altering the shape and strength of the potential as well as including the effects of a distribution of such inhomogeneities. However, our object has only been to show that the presence of physically reasonable lateral fields produces a significant distinction between the behavior of two-dimensional Bose and Fermi systems of the kind observed experimentally.

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[7]It is interesting to note that the same expressions are obtained for a potential $V(x,y)=Kx$ if ξ is replaced by $\beta V(L,y)$, where L is the length of the system.