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Energy spectrum and specific heat of a two-dimensional interacting Bose gas*

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The specific heat and the elementary excitation spectrum of a hard-sphere two-dimensional Bose gas is calculated assuming that only collective modes contribute to the thermodynamical properties of the fluid, at low temperatures. The calculation is based on the self-consistent-field approximation method with only one parameter adjustable, the hard-core diameter. The theoretical curve of the specific heat fits very well the experimental data of the ^{4}He film at density of 0.0279 Å^{-2}. The energy spectrum obtained in this work, which is qualitatively similar to the three-dimensional version, indicates the possibility of occurrence of superfluidity in such films.

I. INTRODUCTION

Monolayers of ^{4}He have been extensively studied in the last few years but the use of grafoil as the substrate has remarkedly improved the reproducibility of the experimental results in such a way that the main theoretical understanding of those films is probably close at hand. For a very extensive review of the subject the reader is referred to the excellent work of Dash and Schick.[1] Experimentally, the specific heat of those films has been shown to be their most interesting feature, its deviation from the bulk ^{4}He results being remarkable in many aspects. It is also to be mentioned that superfluidity has been observed in those films.[2] One of the first measurements of the specific heat of helium films close to the λ point was performed by Frederikse[3]; it showed a progressive rounding of the anomaly with decreasing thickness, together with a shift of the maximum toward lower temperatures. Moreover, the heat capacity of thin films goes as T^2 for very low temperatures and this behavior suggested the possibility of formation of solid layers,[4] a two-dimensional Debye solid. Typically, the measured specific heat of thin ^{4}He films may be summarized as follows (for reference we consider the data corresponding to a density $\rho = 0.0279$ Å^{-2} on grafoil): (a) a "high-temperature" region, above 3 °K, where the system is weakly interacting and can be treated theoretically by virial-expansion calculations[5]; (b) as the temperature is decreased the specific heat increases and goes through a round maximum at about $T_c = 1.2$ °K. This increase in the specific heat for $T > T_c$ can again be understood from virial expansions and is a direct consequence of the hard-sphere part of the interaction, as shown in Ref. 5; (c) a low-temperature region exhibiting a two-dimensional phononlike behavior T^2.

The T^2 behavior at low temperature suggests the existence of a phononlike spectrum for the excitation of the liquid. The reason for sticking to a liquid model at low temperatures is necessary if one intends to explain the observed superfluidity of the films. It seems clear[6] that even a monolayer should exhibit superfluidity if it were not for the strong interactions with the substrate occurring in real experiments.

In 1956, Penrose and Onsager[7] showed that Bose-Einstein condensation can occur in infinite systems of interacting bosons, reinforcing the picture that the superfluidity of liquid helium was dependent, in an essential way, on the microscopic occupation of the $\vec{k} = 0$ state. Later, two works[8] proved that Bose-Einstein condensation could not exist at finite temperatures in two-dimensional systems.

It seems by the two preceding paragraphs that we are faced with a paradox; on the one hand, the experimental evidence of the superfluidity of thin films, and on the other hand, the exclusion of a Bose-Einstein condensation for finite temperatures in two-dimensional systems.

Is the existence of a Bose-Einstein condensation really necessary for superfluidity to occurs? Let us first transcribe from a famous book by Landau and Lifshitz on *Statistical Physics*[9]: "To avoid misunderstanding we must emphasize that these particles ($\vec{k} = 0$) cannot be identified with the 'superfluid part' of the liquid. Apart from the fact that such an identification could have no justification, its incorrectness is seen from the fact that at absolute zero the whole of the liquid becomes superfluid, whereas by no means all its particles have zero momentum."

Superfluidity of a liquid is *really* a consequence of the spectrum of elementary excitations: the absence of excitations with ratio $E(q)/q$ below a certain minimum value.[10] Moreover, since the existence of a spectrum of this kind does not require the occurrence of a Bose-Einstein condensation, as shown by several authors for three-dimensional systems,[11] the theorems proving the absence of a condensation for the layers should not upset the picture of two-dimensional superfluids.[12]

The purpose of this paper is to calculate the specific heat of a layer of liquid helium in the low-temperature region. For this purpose we first calculate the excitation spectrum for a two-dimensional hard-sphere Bose gas,[13] in a self-consistent-field approximation using the same techniques employed successfuly for three-dimensional systems.[14] From the excitation spectrum the specific heat is calculated and the transition temperature, for a certain density, estimated using Landau's approach. Our results fit very nicely the experimental data, with only one adjustable parameter, the hard-sphere radius. It is shown that the energy spectrum, structure factor, pair-correlation function, and other properties of the system are very similar to those of bulk helium.

In Sec. II a brief account of the self-consistent-field approximation is presented in order to obtain the elementary excitation spectrum of a two-dimensional hard-sphere Bose system in Sec. III. Section IV is devoted to a calculation of the specific heat of the system together with a discussion of superfluidity on thin films. In Sec. V we present concluding remarks.

II. SELF-CONSISTENT-FIELD APPROXIMATIONS

Recently, Singwi *et al.*[15] developed a new treatment for the susceptibility of a many-electron system. In their theory the short-range correlations, responsible for the local-field correction are calculated in a self-consistent way, and the susceptibility becomes a functional of the self-correlation function $g(\vec{r})$.

The ansatz, in the semiclassical version replaces the two-particle distribution function in the Liouville equation for the one-particle distribution function $f(\vec{r},\vec{p}\,|\,t)$ by

$$f_2(\vec{r},\vec{p};\vec{r}',\vec{p}'\,|\,t)=f_1(\vec{r},\vec{p};t)f_1(\vec{r}',\vec{p}';t)g(\vec{r},\vec{r}';t), \tag{2.1}$$

thus making it possible to close the hierarchy of equations. In the zero-frequency and infinite-wavelength limit $g(\vec{r},\vec{r}';t)$ can be written

$$g(\vec{r},\vec{r}';t)=g(\vec{r}-\vec{r}')+a\rho\frac{\partial}{\partial\rho}g(\vec{r}-\vec{r}'), \tag{2.2}$$

where $g(\vec{r}-\vec{r}')$ is the equilibrium static pair-correlation function, ρ is the density of the fluid, and a is treated as a parameter which guarantees the fulfillment of the compressibility sum rule.

On the other hand we have the Fourier transform of $g(\vec{r})-1$, which is $(1/\rho)[S(\vec{q})-1]$, where the structure factor $S(\vec{q})$ is related to the susceptibility of the fluid through the well-known relation

$$S(\vec{q})=-\frac{1}{\pi\rho}\int_0^\infty \operatorname{Im}\chi(\vec{q},\omega)d\omega. \tag{2.3}$$

With the ansatz above, the susceptibility of the fluid becomes

$$\chi(\vec{q},\omega)=\chi_0(\vec{q},\omega)/[1-\psi(\vec{q})\chi_0(\vec{q},\omega)], \tag{2.4}$$

where $\chi_0(\vec{q},\omega)$ is usually the screened susceptibility which, in our case, is taken to be the free-gas susceptibility with the same mass, density, and statistics as the one under consideration. $\psi(\vec{q})$ is the self-consistent potential, given by

$$\psi(\vec{q})=\phi(\vec{q})+\left(1+a\rho\frac{\partial}{\partial\rho}\right)\times\int\frac{d\vec{q}'}{(2\pi)^3}\frac{\vec{q}\cdot\vec{q}'}{q^2}\phi(\vec{q}')[S(\vec{q}-\vec{q}')-1]. \tag{2.5}$$

Here $\phi(\vec{q})$ is the bare particle-particle interaction. The susceptibility is now a functional of $S(\vec{q})$ and the scheme is thus closed.

The zero-temperature susceptibility of a noninteracting Bose gas is simply

$$\chi_0(\vec{q},\omega)=2\rho\epsilon(\vec{q})/[(\omega+i\eta)^2-\epsilon(\vec{q})^2], \tag{2.6}$$

where $\epsilon(\vec{q})=\hbar^2q^2/2m$ is the free single-particle energy, and m is the mass of Bose particle. The pole of the full susceptibility $\chi(\vec{q},\omega)$ is given by the relation

$$(\omega+i\eta)^2-\epsilon(\vec{q})^2-2\rho\epsilon(\vec{q})\psi(\vec{q})=0. \tag{2.7}$$

Fixing $\vec{q}$, the solutions for ω define the energy of the excitation $E(\vec{q})$, leading to the spectrum of the density fluctuation

$$E(\vec{q})=\{\epsilon(\vec{q})^2+2\rho\epsilon(\vec{q})\psi(\vec{q})\}^{1/2}. \tag{2.8}$$

On the other hand, $E(\vec{q})$ is related to $S(\vec{q})$ by Feynman's expression

$$E(\vec{q})=\epsilon(\vec{q})/S(\vec{q}), \tag{2.9}$$

which can be readily obtained by integrating the right-hand side of Eq. (2.3). To evaluate $E(\vec{q})$, or equivalently $S(\vec{q})$ it is needful to obtain $\psi(\vec{q})$.

$\psi(\vec{q})$, in our two-dimensional system, is connected to the equilibrium pair-distribution function $g(r)$ through Eq. (2.5) by the relation

$$\psi(\vec{q})=-\frac{2\pi}{q}\int_0^\infty dr\,\frac{d\phi(r)}{dr}rJ_1(qr)\times\left(g(r)+a\rho\frac{\partial}{\partial\rho}g(r)\right). \tag{2.10}$$

Here $\phi(\vec{r})$ is the inverse Fourier transform of $\phi(\vec{q})$,

$$\phi(\vec{r})=\frac{1}{2\pi}\int e^{-i\vec{q}\cdot\vec{r}}\phi(\vec{q})\,d^2q, \tag{2.11}$$

and $J_1(x)$ is the Bessel function of the first kind of order 1.

For a hard-disk interaction we follow the spirit of Ref. 12, by putting

$$\frac{d\phi(r)}{dr}=\lim_{\lambda\to\infty}(-\lambda)\delta(r-r_c), \tag{2.12}$$

where r_c is the hard-disk diameter, we get for $\psi(\vec{q})$ the following expression:

$$\psi(\vec{q})=\lim_{\lambda\to\infty}\frac{2\pi}{q}\lambda r_c J_1(qr_c)\left(g(r_c)+a\rho\frac{\partial}{\partial\rho}g(r_c)\right). \tag{2.13}$$

The term in large parentheses on the right-hand side of Eq. (2.13) is a constant and must be of the order λ^{-1} so that $\psi(\vec{q})$ remains finite for all q's. This is consistent with the fact that as $\lambda\to\infty$ the pair-correlation function must approach to zero as r tends to r_c from above. Then, we must rewrite $\psi(\vec{q})$ as

$$\psi(\vec{q})=\frac{2\pi}{q}\frac{B}{8mr_c}\left(\frac{r_0}{r_c}\right)^2 J_1(qr_c), \tag{2.14}$$

where $2r_0$ is the average particle distance. The factor $(1/8mr_c)(r_0/r_c)^2$ was introduced for future convenience, and the constant B is a function of the density of the system. It is calculated by imposing that $g(r_c)=0$, i.e.,

$$\lim_{r\to r_c}g(r)=\lim_{r\to r_c}\left\{1+\frac{1}{2}\left(\frac{r_0}{r_c}\right)^2\int_0^\infty dx\,x\,J_0\left(\frac{xr}{r_c}\right)\left[\left(1+\frac{BJ_1(x)}{x^3}\right)^{-1/2}-1\right]\right\}=0. \tag{2.15}$$

Here $J_0(x)$ is the Bessel function of order zero. Equation (2.15) may be rewritten

$$1+\tfrac{1}{2}(r_0/r_c)^2F(B)=0, \tag{2.16}$$

where $F(B)$ is the value of the x integral. Now for every ratio $(r_0/r_c)^2$, B is determined so that $g(r_c)=0$. A plot of B vs r_c/r_0 and $F(B)$ vs B is given in Fig. 1.

III. ELEMENTARY EXCITATION SPECTRUM

Inserting the effective potential $\psi(\vec{q})$ obtained in Eq. (2.14) into Eq. (2.9) we may write the following expression for $E(\vec{q})$:

$$E(\vec{q})=\frac{\hbar^2q^2}{2m}\left(1+\frac{BJ_1(qr_c)}{(qr_c)^3}\right)^{1/2}. \tag{3.1}$$

In Fig. 2 several plots of $E(\vec{q})$ vs qr_c are shown as function of B. We note the rotonlike region for values of $B>120$. Note also the progressive depletion of the minimum of $E(\vec{q})$ as B increases, in the same way as for the three-dimensional system. Using now the Feynman expression and the results obtained for $E(\vec{q})$ we calculated $S(\vec{q})$. The results are shown in Fig. 3 for various values of B.

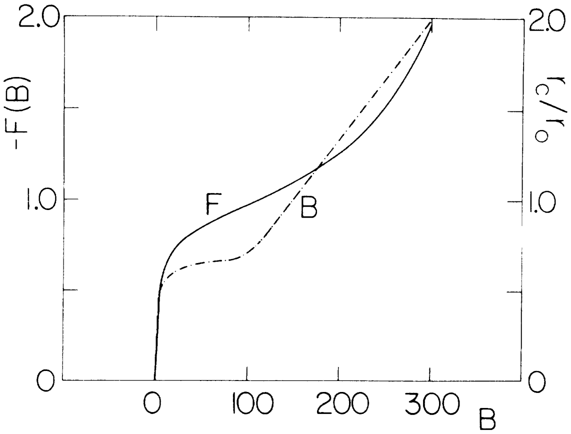

FIG. 1. In the left-hand ordinate we give a plot of $F(B)$ as a function of B, and in the right-hand ordinate we plot B as a function of r_c/r_0, leading to the solutions of Eq. (2.16) of the text.

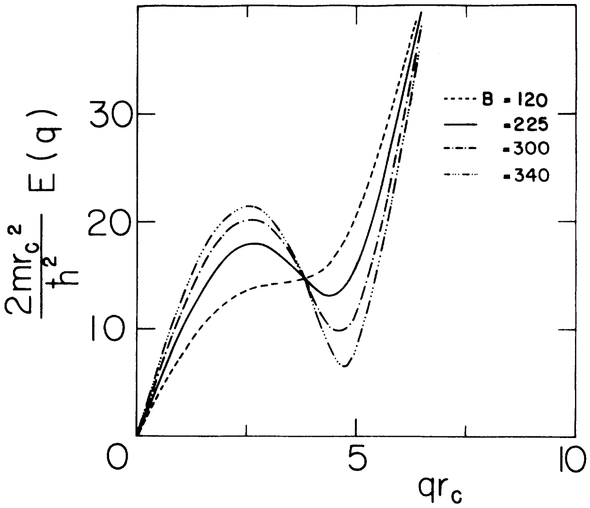

FIG. 2. Energy spectra at several values of the parameter B showing the onset of a rotonlike dip as the density or equivalently B is increasing.

The pair-correlation function $g(r)$ is given by

$$g(r)=1+\frac{1}{2\pi\rho}\int_0^\infty dq\, qJ_0(qr)[S(q)-1]. \tag{3.2}$$

In Fig. 4 we show a typical plot of $g(r)$ vs r/r_c.

Once the energy spectrum is obtained we may calculate the sound velocity v_s which is defined as

$$v_s=\lim_{q\to 0}\frac{E(q)}{q}=\left(\frac{B}{2}\right)^{1/2}\frac{\hbar}{2mr_c}. \tag{3.3}$$

If we insert B and r_c into this expression we finally find for the sound velocity the value $v_s = 157$ m/sec.

To obtain the roton effective mass m^* we expand the energy $E(\vec{q})$ around the wavelength q_0 corresponding to the roton minimum

$$E(q)=E(q_0)+\frac{\hbar^2}{2}(q-q_0)^2\left(\frac{d^2E}{dq^2}\right)_{q=q_0} \tag{3.4}$$

$$=E(q_0)+\frac{\hbar^2(q-q_0)^2}{2m^*},$$

thus defining m^*. We now change variables qr_c $=x$ and get using the fact that $(dE/dq)_{q=q_0}=0$,

$$\frac{m}{m^*}=\frac{1}{S(x_0)}-\frac{x_0^2S''(x_0)}{2|S(x_0)|^2}. \tag{3.5}$$

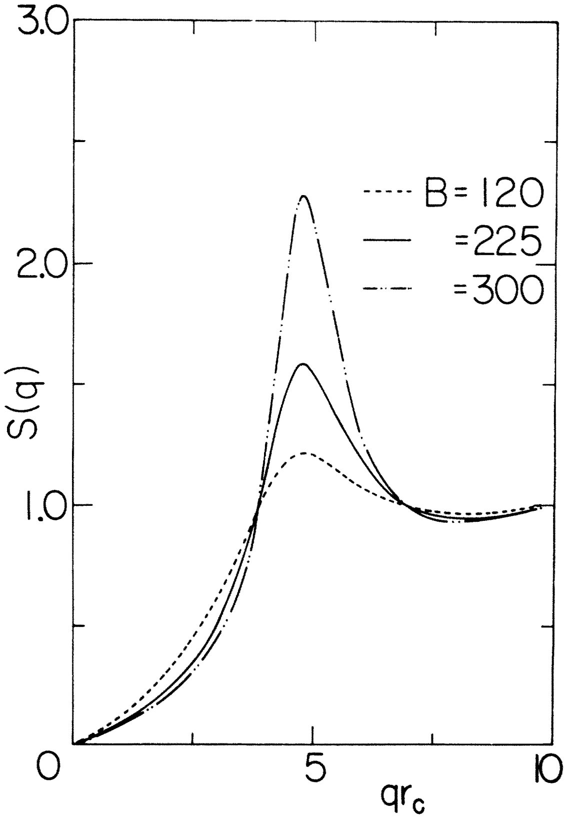

FIG. 3. Structure factor function $S(q)$ obtained from the Feynman expression for various values of the parameter B.

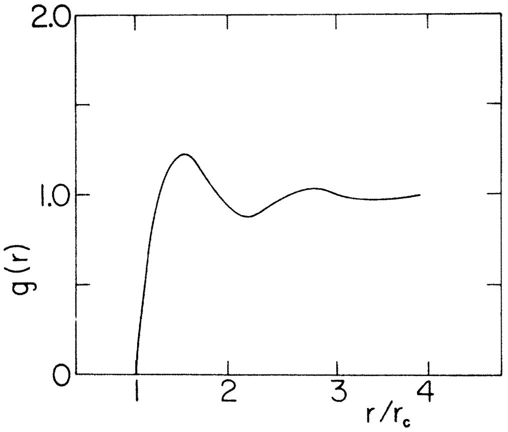

FIG. 4. Pair-correlation function $g(r)$ obtained from Eq. (3.2) at $B = 225$ and at the density of the two-dimensional ^{4}He of 0.0279 Å^{-2}.

IV. SPECIFIC HEAT

The specific heat of a film will be calculated in the low-temperature region, following Landau's approach, i.e., by considering the liquid as replaced by a gas of elementary excitations. Once the elementary excitation energy spectrum for the liquid is known, the calculation of the specific heat is straightforward.

For the purpose of obtaining a quantitative results we shall make use of the energy spectrum calculated in Sec. III for values of $B = 225$ and the areal density $\rho = 0.0279$ Å^{-2}.

The phonon contribution to the specific heat is calculated by extending the upper limit of the $\vec{p}$ integral, in the density of free energy F_{ph} to infinity

$$F_{\text{ph}}=\frac{kT}{(2\pi\hbar)^2}\int_0^\infty \ln(1-e^{-v_s p/kT})d^2p$$

$$=-0.19\frac{k^3T^3}{\hbar^2v_s^2}. \tag{4.1}$$

Substituting into Eq. (4.1) the expression given by Eq. (3.3) for the sound velocity v_s we find the specific heat

$$\frac{C_{\text{ph}}}{Nk}=6.95\, r_c^2T^2/\rho B, \tag{4.2}$$

which is proportional to the square of the temperature.

The roton contribution is obtained by noting that the gas exhibits energy spectrum of the form

$$E(\vec{q})=\Delta+\hbar^2(q-q_0)^2/2m^*, \tag{4.3}$$

where q_0 is the point of minimum of the function $E(q)$, $\Delta = E(q_0)$, and m^* is the roton effective mass. In our calculation, $m^* = 0.2m$.

Since the roton energy includes the quantity Δ, which is large compared with kT at temperatures sufficiently low for the roton gas we are considering, this gas may be described by Maxwell-Boltzmann statistics. Hence the roton contribution to the specific heat is given by

$$\frac{C_r}{Nk} = \left(\frac{m^* kT}{2\pi\hbar^2}\right)^{1/2} q_0 e^{-\Delta/kT}\left[\frac{3}{4} + \frac{\Delta}{kT} + \left(\frac{\Delta}{kT}\right)^2\right]. \tag{4.4}$$

We see that the temperature dependence of the roton part of the specific heat is essentially exponential. At sufficiently low temperatures the roton contribution is smaller than that of the phonon part. Close to the transition temperature the roton part is much greater than the phonon one.

The theoretical curve is plotted in Fig. 5 together with the experimental data. The value of B was chosen to best fit the data. Ours results are in qualitative agreement with those obtained by Miller and Woo.[16]

Assuming that the system under consideration is a superfluid at sufficiently low temperature the transition temperature superfluid-normal liquid may be estimated, again following Landau, by considering the mass density carried by the thermal excitations present at some temperature. When this density is the same as that of the fluid, we assume that the transition temperature is reached since the fluid becomes totally normal, no superfluid component remains. For bulk ^{4}He the transition temperature estimated in this way ($T_\lambda = 2.8$ °K) is larger than the observed one, $T_\lambda = 2.19$ °K.

For a two-dimensional system let us imagine that the "quasiparticle gas" moves as a whole with respect to the liquid, with a translational velocity $\vec{v}$. Hence the total momentum of the gas per unit area is

$$\vec{P} = \int \vec{p}\, n(E - \vec{p}\cdot\vec{v})\, d^2p, \tag{4.5}$$

where $n(E - \vec{p}\cdot\vec{v})$ is the distribution function. Assuming that the velocity $\vec{v}$ is small, we find

$$\vec{P} = -\vec{v}\frac{1}{4\pi\hbar^2}\int_0^\infty dp\, p^3 \frac{dn(E)}{dE}. \tag{4.6}$$

The effective mass per unit area of the gas is determined by the proportionality coefficient between the momentum $\vec{P}$ and the velocity $\vec{v}$ in Eq. (4.6). Thus we may find the phonon part ρ_n^{ph} of the mass density ρ_n of the normal liquid

$$\rho_n^{\text{ph}} = 0.57(kT)^3/\hbar^2 v_s^4. \tag{4.7}$$

To calculate the roton part of ρ_n we note that, since the rotons can be described by a Maxwell-Boltzmann distribution, $dn(E)/dE = -n(E)/kT$ and from Eq. (4.6),

$$\rho_n^r = \frac{\hbar q_0^3}{2}\left(\frac{m^*}{2\pi kT}\right)^{1/2} e^{-\Delta/kT}. \tag{4.8}$$

At low temperatures the roton part of ρ_n is comparable with the phonon part, but at higher temperatures the roton part predominates. Then the transition temperature T_c can be approximately obtained by putting $\rho_n^r/\rho = 1$. Here ρ is the mass density of the fluid. The resulting estimation for T_c is 1.5 °K. If this result is adjusted downwards by the ratio of the observed bulk temperature of the λ point to the estimated bulk transition temperature of ^{4}He, we obtain a T_c of 1.2 °K.

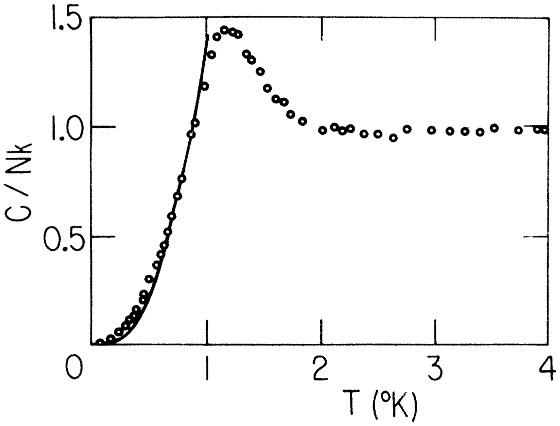

FIG. 5. Specific heat of a two-dimensional ^{4}He at density of 0.0279 Å^{-2}. The specific heat varies as T^2 at low temperatures. The circles correspond to the experimental data according to Ref. 1. The solid line is the theoretical result obtained in this work at the parameter $B = 225$.

V. CONCLUSIONS

From the collective modes spectrum of a two-dimensional hard-sphere Bose system, we obtained a theoretical curve for the specific heat of helium films with one adjustable parameter, the hard-sphere radius.

For a hard-sphere diameter $r_c \simeq 5$ Å, the theoretical curve fits very well the experimental data of ^{4}He films at the density of 0.0279 Å^{-2}. The spectrum of elementary excitations is compatible with a superfluid behavior of those films.

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[12] At this point we must stress than superfluidity is not a necessary support for the results presented in this work. Our calculations are based on the assumption that only collective modes are important thermodynamically, in the low-temperature limit. To establish superfluidity we have to prove that no other kind of low-energy excitation can occur. We, nevertheless, expect that superfluidity is present in the films because the existence of phonons as excitations on a Bose system should prevent the occurrence of single particle low-lying excitations, as discussed in Ref. 10.

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