FEATURES OF THE CONFINED HELIUM ^{4}He SPECIFIC HEAT NEAR THE λ-POINT IN PLANE MESOSCALE PORES

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UDC 536.764
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The influence of the finite-size effect on the liquid helium specific heat and a shift of the transition temperature are theoretically examined for the case of planar confinement. A considerable growth of the specific heat manifests itself at a new transition temperature which is calculated in the context of the present geometric conditions. The analytic results are found to be in fair agreement with a number of experiments where ^{4}He films were ranged in thickness from 48 nm to 57 μm that can be referred as the mesoscale values. The contributions to the shift of the transition temperature caused by the gravitation effect and by the finite-size effect are examined.

1. Introduction

Many systems of experimental and theoretical interest, such as thin films, silicon wafers, interfaces, biomembranes, synaptic clefts, etc., have a reduced planar geometry. The physical properties of the systems undergoing critical phenomena and phase transitions are essentially influenced by spatial limitations. Experimental studies of such systems that can exhibit size-dependent second-order phase transitions provide principal results for the verification of the theory of finite-size effects. The various peculiarities of confined systems represent a long-standing interest in the physics of liquids. Despite the active experimental studies, there is a steady demand for more precise tests of the existing theoretical predictions. New horizons in this field would be opened upon conducting the new series of experiments in fundamental physics [1] that are developed for the realization on the International Space Station (ISS).

A better understanding of the effect of spatial limitation and finite-size-induced phenomena cannot be reached without the analysis of the fundamental experiments with confined helium. Amongst the pioneer studies of the spatial-limitation effect, we mention the helium experiment conducted by Chen and Gasparini [2] more than the quarter of a century ago. More sophisticated measurements for films were reported later on in (see, e.g., [3]).

Conducting the experiments with confined helium is rather difficult due to a number of complicated circumstances. In such Earth-based measurements, the characteristic size of a sample is supposed to be small enough in order to neglect the influence of gravity. On the Earth orbit, such a limitation vanishes. On the Earth in the measurements of specific heat at the helium lambda point, the resolving power is limited due to the competition between the finite-size effect and the pressure dependence of the transition temperature (the gravity effect). Therefore, it is important to be sure that just the finite-size effects cause the observed rounding and shift of the specific heat maximum. A strong desire of researchers to eliminate the influence of the gravity effect became a reason of performing the extremely precise experiment on the near-Earth orbit. Most reliable data on the confined helium heat capacity were reported by the group of J. Lipa during the last decade [4, 5].

In this paper, we are going to present the comparative study that is focused on the consideration of the temperature dependence of the liquid helium heat capacity near the λ-point. The system to be considered hereafter has a reduced geometry in the form of a plane-parallel layer with the typical thickness from a few hundreds of angstroms and over. Such a range of sizes can be associated with the upper border of the nanoscale and a substantial part of the microscale and can be referred to as the mesoscale. Films represent an example of the so-called “well-defined geometry” that allows one to perform direct analytic calculations. The verification of the validity of the theoretical results proposed here is conducted by comparing them with high-resolution experimental data [3—6].

2. Temperature and Size Dependence of Specific Heat

The fluctuation effects must be taken into account in order to develop a reliable prediction of system’s behavior near a second-order phase transition. A singular growth of the correlation length ξ can be realized only for spatially infinite systems. The expression that is predicted by the fluctuation (scaling) theory [7] reads as

$$\xi = \xi_0 \tau^{-\nu}. \tag{1}$$

Here, ξ_0 is the amplitude of the correlation length, $\tau = |T - T_c|/T_c$ is the temperature variable, and ν is the correlation length exponent. To use Eq. (1) for systems of experimental interest, it is necessary to satisfy the condition that assumes that the linear size of a system, L, is much larger than the correlation length ξ. Physical properties and, in particular, correlation properties (see, e.g., [8, 9]) of finite-size systems are different from those of the bulk systems. A system can be considered as finite-sized near the critical point or the phase transition point if its characteristic linear size becomes comparable with the correlation length ξ. It is possible to examine the characteristics of the liquid systems with well-defined reduced geometry (film, cylinder, bar, sphere, or cube) in terms of the pair-correlation function G_2 and the associated correlation length ξ.

Let us consider the geometry of a thin liquid system in the form of a plane-parallel layer $D \times D \times H$. The layer thickness H is supposed to be much smaller than the distance D in XY directions. We will study the region where the value of correlation length ξ becomes comparable or even larger than H but still much smaller than D. The pair correlation function G_2 for the planar geometry was deduced by applying the Helmholtz operator procedure (see, e.g., [9, 10]). One can get the expression for G_2 in the form:

$$G_2(x,y,z) = \frac{1}{2}\pi H \sum_{n=0}^{\infty} (1-(-1)^n) \times$$

$$\times K_0 \left[\sqrt{x^2+y^2} \left(\xi^{-2} + \frac{n^2\pi^2}{H^2} \right)^{1/2} \right] \cos\left(\frac{n\pi}{H} z \right). \tag{2}$$

Here, $K_0(u)$ is the cylindrical McDonald function. Because of a non-exponential shape of G_2, the correlation length of density fluctuations can be defined as $\xi = (M_2)^{1/2}$, where M_2 denotes the normalized second spatial moment of G_2 . Following the method described elsewhere, it is possible to get the dominating component of the correlation length in the XY-plane:

$$\xi^* = \xi_0 \left[\left(\left(\frac{\pi\xi_0}{H} \right)^{\frac{1}{\nu}} + 1 \right) \tau + \left(\frac{\pi\xi_0}{H} \right)^{\frac{1}{\nu}} \right]^{-\nu}. \tag{3}$$

The correlation length ξ^* of a thin liquid film depends, naturally, not only on the temperature variable τ but also on the thickness H of a layer. One can suggest that the expression

$$\tau^* = \left(\left(\frac{\pi\xi_0}{H} \right)^{\frac{1}{\nu}} + 1 \right) \tau + \left(\frac{\pi\xi_0}{H} \right)^{\frac{1}{\nu}}$$

represents the new temperature variable that governs the near-critical behavior of the liquid film system. Based on the current results, it would be possible to estimate the linear size of ordered clusters, which is, obviously, appears to be a function of τ and H. Near the phase-transition point, this size can essentially exceed the distance associated with the direct intermolecular interaction and become macroscopic. The film geometry, as well as the cylindrical one [10], makes the correlation properties anisotropic. Consequently, it is believed that, at the achievement of the new critical temperature of a finite-size liquid $T_c^*(H) < T_c$ in a sample of the planar geometry, a considerable growth of the correlation length ξ^* along the unrestricted direction of XY-plane might exist.

In order to verify the validity of the theoretical results proposed here, it is important to have a possibility to compare them with high-resolution experimental data. In view of the fact that most of the data [3—6], which are available up to this date, were derived from the studies of the heat capacity of confined ^{4}He, it is necessary to obtain an analytic expression particularly for this property.

Regardless of the geometry, the relationship between the heat capacity C and correlation length ξ is given by the expression (see, for example, [11]):

$$C \sim \xi^{\alpha/\nu}. \tag{4}$$

In fact, near the lambda point of liquid helium, a small negative value of $\alpha \approx -0.026$ was observed in early experiments. Later on, in the recent revision [12] of the most precise microgravity experiment [5] on the near-Earth orbit, it was confirmed to be $\alpha = -0.0127 \pm 0.0003$. However, it is known that the exponent α appears to be positive and close to 0.1 near the liquid-gas critical point. On the other hand, for liquid ^{4}He, the hyperscaling relation $\alpha = 2 - 3\nu$ with ν=0.6705 [13] yields $\alpha/\nu = -0.0172$.

Thus, adopting Eq. (3), by assuming $\xi = \xi^*$, and making an ansatz for Eq. (4), we can write the formula for heat capacity $C_{\rm plan}(\tau, H)$ in case of planar geometry as

$$C_{\rm plan}(\tau, H) \sim \left[\left(\left(\frac{\pi\xi_0}{H}\right)^{\frac{1}{\nu}} + 1\right)\tau + \left(\frac{\pi\xi_0}{H}\right)^{\frac{1}{\nu}}\right]^{-\alpha}, \quad (5)$$

with H being the film thickness. The amplitude of the correlation length ξ_0 for helium is equal to 0.36 nm below and is by 0.143 nm above the transition temperature.

Next, we will examine the limiting cases, namely $H \to \infty$ and τ=0. Equation (5) appears reasonable and leads to the expected expressions

$$C_{\rm plan}(\tau, H \to \infty) \sim \tau^{-\alpha}, \quad (6)$$

which obviously demonstrate the classical bulk behavior, and

$$C_{\rm plan}(\tau = 0, H) \sim (\pi\xi_0)^{-\frac{\alpha}{\nu}} H^{\frac{\alpha}{\nu}}. \quad (7)$$

Thus, Eq. (7) shows that the heat capacity remains finite in the planar geometry at T_λ (τ=0) if $H < \infty$. Nevertheless, as seen from Eq. (5), $C_{\rm plan}(\tau, H)$ grows up to its limit at some temperature below T_λ in the region of negative τ. These two simple tests represented by Eq. (6) and Eq. (7) can be considered as an indication of the agreement of the proposed results with the finite-size scaling predictions [14] and the renormalization-group calculations [15]. However, it seems that the 2D-crossover for the planar confinement, i.e. the case where $H/\xi_0 \to 1$, cannot be checked in terms of Eq. (5). The other test to be discussed henceforward involves a temperature shift in the heat capacity maximum depending on the film thickness H.

The heat capacity $C_{\rm plan}(\tau, H)$ of liquid ^{4}He under the planar confinement of 5039 and 1074 Å thick, which corresponds to data [3], is presented in Fig. 1 versus the reduced temperature τ on a semilogarithmic scale. The temperature dependence of the heat capacity in Fig. 1 has a finite peak, although it is reminiscent to the distinctive lambda-shape. However, in the actual experiments, the heat capacity peak is considerably rounded. Nevertheless, the temperature location of the peaks in all cases is moderately close to experimental ones. From Fig. 1, one can see that, whilst the thickness H becomes larger, the heat capacity $C_{\rm plan}(\tau = 0, H)$ at T_λ increases in contrast to the temperature shift in the heat capacity maximum that decreases.

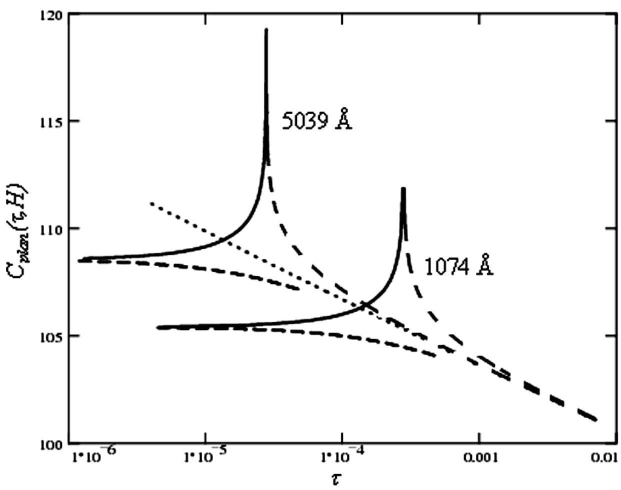

Fig. 1. Dependence of the confined helium heat capacity $C_{\rm plan}(\tau,H)$ (arbitrary units) on the reduced temperature τ for film samples on the semilogarithmic scale [according to Eq. (5)]. The higher curve represents the 5039-Å film, and the lower one does the 1074-Å one

3. Discussion

Let us find a new size-dependent value of the critical temperature $T_c(H)$ in contrast to its unique bulk value $T_c(\infty)$ for the unconfined liquid. It is possible to show that the amplitude of the critical temperature shift in a reduced geometry can vary depending on the choice of a particular physical property. However, because of Eq. (4), one can suggest that the maxima for C and ξ in finite-size systems take place at the same temperature. Equation (5) for the heat capacity keeps well-defined parameters that are related to the planar geometry of the system. It makes possible an easy comparison with the data of corresponding experiments. Proceeding forward, following the procedure described elsewhere, and rearranging Eq. (5), we can find the new transition temperature $T_c(H)$ that indicates a location of the heat capacity maximum in the liquid film system:

$$T_c^*(H) = T_c[1 + (\pi\xi_0/H)^{1/\nu}]^{-1}. \quad (8)$$

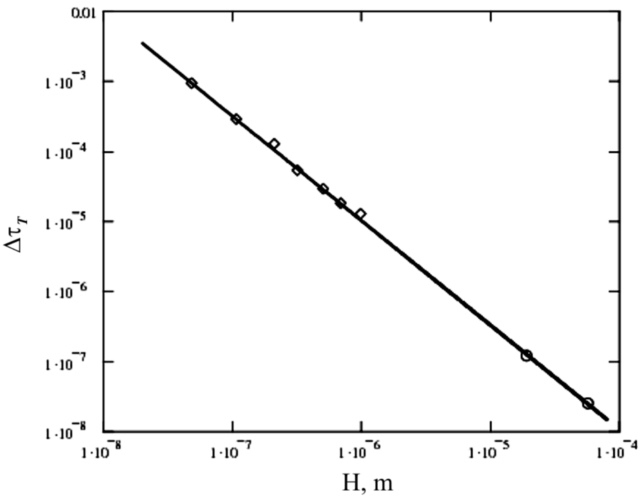

Fig. 2. Dependence of the shift of the transition temperature $\Delta\tau_{\mathrm{t}}$ on the film thickness H on the log-log scale [according to Eq. (9)] — solid line. The slope of the plot corresponds to $\nu = 0.6705$. $\diamond$ represents the experimental data of the Gasparini Group [3], and $\circ$ represents the experimental data of the Lipa Group [4]

The experimental data on liquid helium films [3,4] and the current analytic estimations are combined in the table. There, $\Delta\tau_{\mathrm{e}} = (T_\lambda - T_m)/T_m$ represents the temperature shifts of the helium heat capacity maximum T_m, that is observed experimentally, from its bulk value $T_\lambda \approx 2.1768$ K (see, e.g., [16]), with the reference that $T_\lambda > T_m$. The shifts $\Delta\tau_{\mathrm{t}}$ were calculated according to the proposed theoretical approach with the help of the expression

$$\Delta\tau_{\mathrm{t}} = (\pi\xi_0/H)^{1/\nu}. \tag{9}$$

Points $\Delta\tau_E$ F1—F7 that belong to the size range below 1 μm represent the experimental results of the Gasparini Group [3]. Points F8 and F9 that are correspond to film thicknesses of 19 and 57 μm represent the results of Lipa Group [11].

Transition temperature shift: Experimental data versus the corresponding theoretical estimates for helium confined in film samples of different thicknesses ranged from 48 nm to 57 μm

Thickness H	Experiment $\Delta\tau_{\mathrm{e}}$	Theory $\Delta\tau_{\mathrm{t}}$	Point's number [Ref.]
483 Å	9.356×10^{-4}	9.337×10^{-4}	F1 [6]
1074 Å	2.860×10^{-4}	2.835×10^{-4}	F2 [6]
2113 Å	1.273×10^{-4}	1.033×10^{-4}	F3 [6]
3189 Å	5.3×10^{-5}	5.594×10^{-5}	F4 [6]
5039 Å	2.920×10^{-5}	2.827×10^{-5}	F5 [6]
6918 Å	1.808×10^{-5}	1.762×10^{-5}	F6 [6]
9869 Å	1.278×10^{-5}	1.037×10^{-5}	F7 [6]
19 μm	1.2×10^{-7}	1.260×10^{-7}	F8 [4]
57 μm	2.5×10^{-8}	2.447×10^{-8}	F9 [4]

Using the data in tabular form [6], it was possible to get information for a more precise judgment (point's set $\Delta\tau_{\mathrm{e}}$ F1—F3, F5—F7 in the Table). In the rest of the cases, the points were taken from the graphs presented in the papers [3, 4] and, consequently, have a lower accuracy. The comparison shows that the theoretical values of $\Delta\tau_{\mathrm{t}}$ in most cases (F1, F2, F5, F6, and F9) underestimate the shift of the new transition temperature by 1.8% on the average.

In Fig. 2, we present the dependence of the transition temperature shift $\Delta\tau_{\mathrm{t}}$ on the film thickness H which varies from 10 nm up to 100 μm on the log-log scale. It is seen that the value of $\Delta\tau_{\mathrm{t}}$ decreases with respect to film's thickness expansion. The slope of the plot on Fig. 2 is $1/\nu\approx1.49$ in agreement with the finite-size scaling theory predictions [17]: $\Delta\tau = aL^{-1/\nu}$, where a is a constant depending on the geometry. In the case considered above, the linear size of a system, L, is treated as the film thickness H. As a result, the expression for the scaling coefficient a reads: $a = (\pi\xi_0)^{1/\nu}$.

Let us discuss some features of the influence of gravitation on the shift of the transition temperature in experiments with confined helium. In the close vicinity of the second-order transition temperature, the compressibility of the liquid demonstrates an anomalous increase. The vertical variation of the λ-transition temperature δT_λ over the height H of a chamber due to the hydrostatic pressure induced by the Earth gravity (the gravity effect) is determined by the expression [18]

$$\delta T_\lambda = \gamma H \tag{10}$$

with the coefficient $\gamma = 1.273\mu$ K/cm. It is assumed that data are unaffected by gravity if $|T - T_\lambda| \geq 10\times\delta T_\lambda$ [19]. Obviously, the gravity effect sets the vertical limit of the size for conducting the confined helium experiments on the Earth. At some point, the shift of the transition temperature induced by gravity can become of the same order of magnitude as the shift due to the finite-size effect [20]. Then the separation of these two actual features becomes complicated. Nevertheless, it is possible to neglect the gravity effect and to simultaneously conduct a reliable study of the finite-size effect at some range of the confining sizes. It has been suggested [21,22] that gravity does not play a major role in the actual terrestrial experiments with the typical confining sizes less than 10 μm.

It is possible to compare the contributions to the shift of the transition temperature caused by the gravity effect (δT_g) and by the finite-size effect (δT_{fs}) by direct calculations. Let us introduce the "effect's ratio" $\Omega(H)=(\delta T_g/\delta T_{fs})\times 100\%$ that gives an opportunity to check which effect dominates for any given film thickness. Taking Eqs. (9) and (10) into account, we get

$$\Omega(H) = \frac{\gamma}{(\pi\xi_0)^{1/\nu}\, T_\lambda} H^{\nu+1/\nu} \times 100\%. \quad (11)$$

Following [19] and adopting Eq. (11), it can be suggested that the confined helium experimental results can be considered as unaltered by the gravity if $\Omega(H) \leq 10\%$. In this context, one can determine the exact value of the maximum film thickness for terrestrial experiments as H_L=50.29 μm. It appears to be 5 times larger than it was proposed in [21,23]. It also indicates a disagreement with the statement [23] that the finite-size effect would be totally obscured by the gravity for 50 μm sample. In most cases of the above-discussed terrestrial experiments, this H_L-condition is satisfied. Furthermore, this calculated limit value H_L correlates well with the fact that the experiment with a 57-μm helium film [4] was conducted in the zero-gravity environment on the near-Earth orbit. It could be interesting to compare data [4] with the experimental study of the same confined system which will be conducted on the Earth. Such ultimate test could give a source of information which could be used for the separate and combined evaluation of the gravity and finite-size effects.

In Fig. 3, we give the dependence of the "effect's ratio" $\Omega(H)$ on the film thickness H according to Eq. (11) on the log-log scale. Thus, the ratio $\Omega(H_1)$=13.7% for H_1=57 μm, the ratio $\Omega(H_2)$=0.9% for H_2=19 μm, and this ratio dramatically decreases to 3.4×10^{-5}% for H_3=0.32 μm. On the other hand, Eq. (11) shows that, for the film thickness 96.47 μm, the gravitation effect and the finite-size effect give the equivalent contributions, $\delta T_g=\delta T_{fs}=2.4\times10^{-8}$ K, to the transition temperature shift and, consequently, can mimic each other.

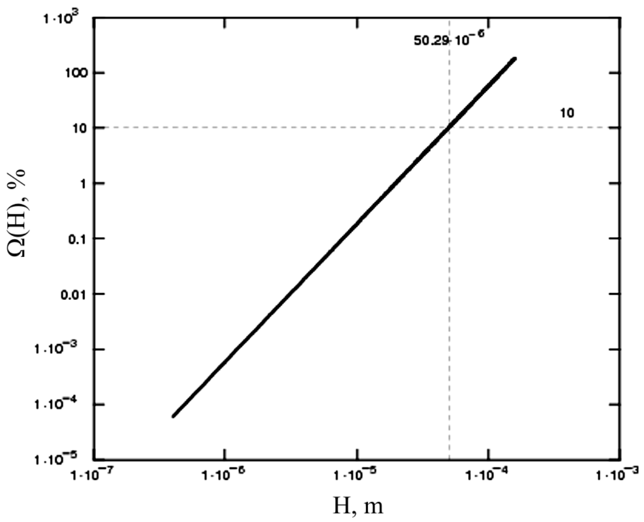

Fig. 3. Dependence of the "effect's ratio" $\Omega(H)=(\delta T_g/\delta T_{fs})\times 100\%$ on the film thickness H [according to Eq. (11)] on the log-log scale. The slope of the plot corresponds to $(\nu+1)/\nu$=2.49

4. Conclusions

We believe that the other static and dynamical physical properties of finite-size systems, which can be represented near the critical point in terms of the scaling relations with correlation length, can be described in the framework of this study. At this point, the current theoretical approach gave the results that are reasonably matched to the confined ^{4}He heat capacity experimental data over the wide range of system's sizes from 50 nanometers up to about 60 micrometers for the film-type geometry.

K. Ch. would like to express the heartiest gratitude to deceased Prof. K. Hamano, Japan, and to thank Prof. L. Bulavin at Kyiv National University, Prof. A. Chalyi at National Medical University, Ukraine, and Profs. K. Kubota and T. Yamamoto at Gunma University, Japan for the numerous useful discussions and the enlightening comments in a course of this research.

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Received 20.09.05

ОСОБЛИВОСТІ ТЕПЛОЄМНОСТІ ОБМЕЖЕНОГО РІДКОГО ^{4}He ПОБЛИЗУ λ-ТОЧКИ В ПЛОСКИХ МЕЗОМАСШТАБНИХ ПОРАХ

К.О. Чалий

Р е з ю м е

Досліджено вплив ефекту просторової обмеженості на теплоємність рідкого гелію ^{4}He та зсув температури λ-переходу для плоскої геометрії системи. Зростання теплоємності відбувається при новій температурі λ-переходу, зсув якої обчислено теоретично. Результати розрахунків виявилися добре узгодженими з даними експериментів, в яких товщина плівок гелію ^{4}He варіювалась в мезомасштабі розмірів від 48 нм до 57 мкм. Вивчено внески в зсув температури λ-переходу, спричинені ефектом просторової обмеженості та дією гравітації.