

doi: 10.15407/ujpe60.09.0885

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68.35.Rh, 61.12.-q,
82.56.Lz**DIMENSIONAL CROSSOVER
AND THERMOPHYSICAL PROPERTIES
OF NANOSCALE CONDENSED MATTER**

The problem going to be discussed is as follows: how results for 3D systems transfer to results for 2D systems and vice versa. Obviously, such a $3D \Leftrightarrow 2D$ dimensional crossover should be smooth and without discontinuities. Here, this problem is studied for a single-component classical liquid in a reduced geometry, namely for slit-like and cylindrical pores with the lower crossover dimensionality $D_{\text{cros}} = 2$ and $D_{\text{cros}} = 1$, correspondingly, which are filled by water molecules. The influence of the $3D \Leftrightarrow 2D$ dimensional crossover on the effective critical exponents α_{eff} , ν_{eff} , γ_{eff} and on the thermophysical properties such as the heat capacity C_V and the isothermal compressibility β_T is investigated.

Keywords: dimensional crossover, confined systems, effective critical exponents, isochoric heat capacity, isothermal compressibility.

1. Introduction

The crossover (transitional) phenomena describing the nature of the critical behavior changes on approaching from a vicinity of one phase-transition point to another one (for example, from the critical to the tricritical point or the crossover events between the Ising model and the Heisenberg model critical behaviors) was discussed in a number of works [1–4].

The dimensional crossover, being a particular case of the general crossover phenomena, usually takes place in systems under confinement of different nature such as low-dimensional magnetic systems, confined fluids, liquid crystals, few-layer graphene, carbone nanotubes, *etc.* [5–14]. The dimensional crossover is governed by the important condition: the correlation length ξ of order parameter fluctuations has to be larger or the same order of magnitude in comparison with a linear size L of system's restricted volume under consideration, i.e. $\xi \geq L$.

A consistent treatment of such kinds of crossover phenomena in nanoscale condensed systems requires to consider the problem of universality classes, for which one has specific conditions of a similar critical behavior of different physical properties. As is known [1, 2, 15–28], these conditions of unversality classes for

bulk systems with $L \gg \xi$ are as follows: (a) the same space dimensionality D ; (b) the same dimensionality (number of components) n of system's order parameter; (c) the same type (short- or long-range) of the intermolecular interaction; (d) the same symmetry of the Hamiltonians (fluctuation part of the thermodynamic potential).

While changing one (or more) of these four conditions, the crossover phenomena may appear in the bulk, as well as in systems under confinement. In the latter case, i.e. in confined systems with $\xi \geq L$, the following additional conditions of universality classes have to be taken into account: (e) the same type (hydrophilic, hydrophobic, or partial wetting) of boundary conditions, (f) the same geometric form of system's limiting surfaces or the same lower crossover dimensionality D_{cros} , (g) the same physical property under consideration [6, 29–31, 43].

The lower crossover dimensionality D_{cros} mentioned in condition (f) means such a dimensionality, which characterizes a limiting volume in the case where its one (two or all three) linear size is approaching a monomolecular thickness [6, 28–30]. Thus, thin films have a form of monomolecular planes with $D_{\text{cros}} = 2$, cylindric pores receive a form of monomolecular wires with $D_{\text{cros}} = 1$, while quantum dots or vesicles have a limiting form of a single molecule with $D_{\text{cros}} = 0$.

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The final condition (g) is considered to be also important because the equations for coordinates of maxima or minima for the physical properties in systems with confinement contain non-universal amplitudes in the scaling laws. Therefore, in contrast to the critical parameters in bulk fluid systems with $L \gg \xi$ having the single critical point, these equations give (a) the analogues of the critical parameters, which are different due to non-universal amplitudes, say, for the heat capacity or the isothermal susceptibility, *etc.*, as well as (b) a non-singular “rounding” critical behavior of the physical properties in a reduced geometry with $\xi \geq L$ [6, 30, 39, 43].

Two types of a dimensional crossover were studied in our previous papers [5, 6, 32–35, 38–41, 43, 44]. The 1st type of a dimensional crossover corresponds to the transition from $3D$ bulk to $3D$ confined liquids. In this case, the dependence of physical properties on thermodynamic variables (temperature, density, pressure, *etc.*) in bulk liquids with linear sizes $L \gg \xi$ converts into the dependence of these properties on linear sizes in confined liquids with $\xi \leq L$. The 2nd type of a dimensional crossover corresponds to the case where a further decreasing of linear sizes in confined liquids could be treated under certain conditions as the change of the spatial dimensionality D (for example, $3D \Leftrightarrow 2D$ crossover in slit-like pores or $3D \Leftrightarrow 1D$ crossover in cylindrical pores). A smooth $3D \Leftrightarrow 2D$ transition of the effective critical exponent ν for the temperature dependence of the correlation length ξ from $\nu = 0.625$ for $D = 3$ to $\nu = 1$ for $D = 2$ is examined for liquids in a reduced geometry in [6, 36].

This paper is aimed at studying the temperature dependence of the isochoric heat capacity C_V and the isothermal compressibility β_T in a single-component classical liquid in a reduced geometry. For this purpose, the effective critical exponents α_{eff} for C_V and γ_{eff} for β_T are calculated with a dimensional crossover of the 2nd type taken into account. For definiteness, all the obtained results are applied to two types of the lower crossover dimensionality $D_{\text{cross}} = 2$ and $D_{\text{cross}} = 1$, correspondingly, in slit-like and cylindrical pores filled by water molecules.

2. Effective Critical Exponent α_{eff} and Heat Capacity C_V

Here, we study the influence of a $3D \Leftrightarrow 2D$ dimensional crossover on the effective critical exponent α and

the heat capacity C_V for confined single-component classical fluids. Numerical results will be obtained for water in a reduced geometry of slit-like and cylindrical pores. Such fluids belong to the universality class of the Ising model and have a scalar order parameter $\varphi = \frac{\rho - \rho_c}{\rho_c}$ related to a deviation of the density ρ from its critical value ρ_c .

In $3D$ case, the isochoric heat capacity in the bulk phase has a “weak” singularity at the critical temperature $T_c(\infty)$:

$$C_V = C_{V0} {}^{3D} \tau^{-\alpha}, \quad (1)$$

where $C_{V0} {}^{3D}$ is the non-universal amplitude of the isochoric heat capacity, $\tau = [T - T_c(\infty)]/T_c(\infty)$ is the temperature variable, and α is the critical exponent, which equals $\alpha_{3D} = 0.125$ for the $3D$ Ising model.

In the $2D$ Ising model, the heat capacity has the well-known Onsager logarithmic divergence:

$$C_V = C_{V0} {}^{2D} \ln \tau, \quad (2)$$

which corresponds to the critical exponent $\alpha_{2D} = 0$ (see, e.g., [2]).

2.1. Effective Critical Exponent α_{eff}

To study a $3D \Leftrightarrow 2D$ dimensional crossover for the critical exponent α_{eff} , we shall use the following formula for any effective critical exponent [6]:

$$\alpha_{\text{eff}} = \alpha_{3D} + \left[\frac{2}{\pi} \arctg(ax - b) - 1 \right] \frac{\alpha_{3D} - \alpha_{2D}}{2}. \quad (3)$$

Here, $x = H/H_{\text{cross}}$ is the dimensionless coordinate of a plane-parallel layer under confinement; H_{cross} is the linear size of fluid’s restricted layer, at which a crossover occurs; a and b are the parameters characterizing the slope and the position of the $3D \Leftrightarrow 2D$ crossover.

Numerical values of the dependence of the critical exponent α_{eff} on the number S of molecular layers, i.e. the slit-like pore’s thickness H or the cylindrical pore’s radius R , are given in Table 1 (see 2nd column).

A $3D \Leftrightarrow 2D$ dimensional crossover of the critical exponent α_{eff} is more expressive within the interval 0.9–3.6 nm of the pore’s thickness H or radius R in confined fluids of a slit-like or cylindrical geometry. Really, the critical exponent α_{eff} is increasing approximately 4 times in this interval: from $\alpha_{\text{eff}}(H = 0.9 \text{ nm}) = 0.03$ to $\alpha_{\text{eff}}(H = 3.6 \text{ nm}) \approx 0.12$ (Fig. 1).

Equation (3) looks similar to the crossover between hydrodynamic and fluctuation regions for the central Rayleigh component of the light-scattering spectrum in the mode-mode coupling version of the dynamical theory of critical phenomena [42, 44].

2.2. Isochoric heat capacity C_V

To calculate the dependence of the heat capacity C_V not only on the temperature but also on the linear sizes of fluid's volume, it is necessary to use, instead of the temperature $\tau = [T - T_c(\infty)]/T_c(\infty)$ in bulk fluids, the following new temperature variable for fluids in a confined geometry [6, 38, 39]:

$$\tau(S, \xi) = (G/S)^{1/\nu} + [1 + (G/S)^{1/\nu}](\xi_0/\xi)^{1/\nu} \text{sign}\tau. \quad (4)$$

Here, G is the geometrical factor depending on the lower crossover dimensionality of fluid's volume (for plane-parallel layers or slit-like pores, $G = \pi$; while, for cylindrical pores, $G = \mu_1 = 2.4048$, where μ_1 is the first zero of the Bessel function $J_0(z)$); $S = L/d_0$ is the number of molecular layers, where L is a linear size of fluid's volume such as layer's thickness or cylinder's radius, and d_0 is the average diameter of a molecule (for water molecules, $d_0 \approx 0.3$ nm).

First, let us consider the 1st type of a dimensional crossover. For relatively large sizes $L \gg \xi$, it is easy to find from Eq. (4) that, due to the factor $S = L/d_0 \gg 1$, the correlation length ξ is approaching its bulk value $\xi = \xi_0 \tau^{-\nu}$. In this case, the inequality

$$\xi_0/\xi \gg (G/S)[1 + (S/G)^{1/\nu}]^\nu \quad (5)$$

is valid. Then the term with (G/S) in (4) may be omitted, and all the physical properties depend on the thermodynamic variables (temperature, density or concentration, pressure, *etc.*). In the opposite case where the inequality

$$S = L/d_0 \gg G(\xi/\xi_0)[1 + (G/S)^{1/\nu}]^{-\nu} \quad (6)$$

takes place, one has the dependence of all the physical properties only on the linear sizes in confined fluids.

The expressions of the isochoric heat capacity in $3D$ and $2D$ confined fluids can be written in the following form, correspondingly, with regard for the new temperature variable (4) and after substituting $\tau(S, \xi)$ instead of the temperature τ into formulae (1) and (2):

$$C_V = C_{V0}^{3D} \left\{ \left(\frac{G}{S} \right)^{\frac{1}{\nu}} + \right.$$

$$\left. + \left[1 + \left(\frac{G}{S} \right)^{\frac{1}{\nu}} \right] \left(\frac{\xi_0}{\xi} \right)^{\frac{1}{\nu}} \text{sign}\tau \right\}^{-\alpha(S)}, \quad (7)$$

$$C_V = C_{V0}^{2D} \ln \left\{ \left(\frac{G}{S} \right)^{\frac{1}{\nu}} + \left[1 + \left(\frac{G}{S} \right)^{\frac{1}{\nu}} \right] \left(\frac{\xi_0}{\xi} \right)^{\frac{1}{\nu}} \text{sign}\tau \right\}. \quad (8)$$

To study explicitly a $3D \Leftrightarrow 2D$ dimensional crossover of the isochoric heat capacity, let us consider the case of relatively small linear sizes S and relatively large correlation lengths ξ , so that inequality (6) is valid. Say, let the number S of molecular layers along the direction of spatial limitation be changing from 10 to 30, while the temperature variable $\tau \approx 10^{-3} - 10^{-4}$, i.e. the correlation length $(\xi/\xi_0) \approx 10^{-2} - 10^{-2.5}$. As a result, the first terms in the braces in (7) and (8) are 10 times larger than

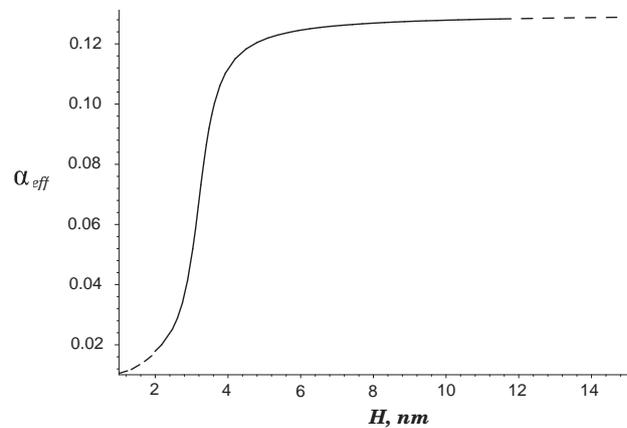


Fig. 1. Critical exponent α_{eff} in a slit-like pore with thickness H filled by water

Table 1. The effective critical exponents in a $3D \Leftrightarrow 2D$ dimensional crossover

S	α_{eff}	ν_{eff}	γ_{eff}
1	$\rightarrow 0$	$\rightarrow 1$	$\rightarrow 1.750$
2	0.029	0.915	1.635
3	0.030	0.913	1.632
4	0.031	0.910	1.628
5	0.032	0.906	1.622
6	0.034	0.900	1.615
7	0.037	0.892	1.604
8	0.041	0.878	1.585
9	0.050	0.853	1.552
10	0.067	0.802	1.483
11	0.095	0.719	1.370
12	0.117	0.655	1.284
13	$\rightarrow 0.125$	$\rightarrow 0.630$	$\rightarrow 1.250$

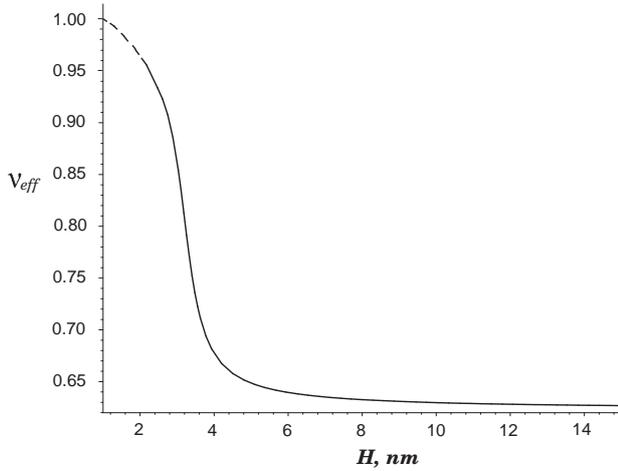


Fig. 2. Critical exponent ν_{eff} in a slit-like pore with thickness H filled by water

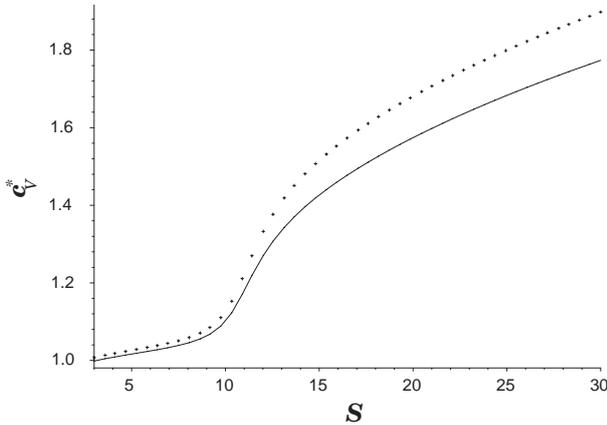


Fig. 3. Heat capacity $C_V^*(S)$

the second ones. Therefore, by omitting the second terms depending on the correlation length, one has the following reduced expressions to study a $3D \Leftrightarrow 2D$ dimensional crossover of the isochoric heat capacity:

$$C_V = C_{V0}^{3D} (G/S)^{-\alpha(S)/\nu(S)}, \quad (9)$$

$$C_V = C_{V0}^{2D} [\ln(G/S)]/\nu(S). \quad (10)$$

The interpolation method proposed in [6] for the effective critical exponents in a $3D \Leftrightarrow 2D$ dimensional crossover may be generalize onto the isochoric heat capacity C_V . This method leads to the following interpolation formula for the heat capacity C_V with regard for formulae (9) and (10) together with formula

(3) and Fig. 2 for the critical exponent $\nu_{\text{eff}}(S)$:

$$C_V = C_{V0}^{3D} \left(\frac{G}{S}\right)^{-\frac{\alpha(S)}{\nu(S)}} + \frac{1}{2} \left\{ \frac{2}{\pi} \arctg(ax - b) - 1 \right\} \times \left\{ C_{V0}^{3D} \left(\frac{G}{S}\right)^{-\frac{\alpha(S)}{\nu(S)}} - C_{V0}^{2D} \frac{\ln \frac{G}{S}}{\nu(S)} \right\}. \quad (11)$$

However, this approach to calculate the heat capacity in a $3D \Leftrightarrow 2D$ dimensional crossover seems to be rather complicated. It is possible to propose a simpler way to find the dependence of the isochoric heat capacity C_V on the linear pore size S (thickness H or radius R) with help of the formula

$$C_V^* = C_V(S)/C_{V0} = (S/G)^{\alpha_{\text{eff}}(S)/\nu_{\text{eff}}(S)}. \quad (12)$$

Equation (12) being similar to (9) uses the effective critical exponents $\alpha_{\text{eff}}(S)$ and $\nu_{\text{eff}}(S)$ in a $3D \Leftrightarrow 2D$ dimensional crossover. The results obtained for the isochoric heat capacity C_V^* in accordance with (12) are presented in Fig. 3.

As is seen, a $2D$ value of the isochoric heat capacity equals 1, and then $C_V^*(S)$ demonstrates a change of the curvature and a S -like growth by factors 1.6 (or 1.5) with increasing the number S of molecular layers to 30, i.e. the thickness H (or radius R) up to 10 nm in slit-like (or cylindrical) pores filled by water (correspondingly, a lower curve in Fig. 3 for slit-like pores with $D_{\text{cros}} = 2$, while an upper curve – for cylindrical pores with $D_{\text{cros}} = 1$).

3. Effective Critical Exponent

γ_{eff} and Isothermal Compressibility β_T

Here, as in the previous section, we continue studying a $3D \Leftrightarrow 2D$ dimensional crossover of the critical exponents and thermophysical properties in single-component fluids in a reduced geometry of slit-like or cylindrical pores filled by water molecules.

3.1. Effective critical exponent γ_{eff}

To investigate the influence of crossover effects on the critical exponent γ_{eff} , we rewrite the formula for any effective critical exponent [6] as follows:

$$\gamma_{\text{eff}} = \gamma_{3D} + \left(\frac{2}{\pi} \arctg(ax - b) - 1 \right) \frac{\gamma_{3D} - \gamma_{2D}}{2}, \quad (13)$$

where $\gamma_{3D} = 1.25$ and $\gamma_{2D} = 1.75$ [2].

Numerical values for the dependence of the critical exponent γ_{eff} on the number S of molecular layers are

presented in Table 1 (see the 4th column). It is worth to mention that the dependence $\gamma_{\text{eff}}(S)$ may be easily calculated as the dependences $\gamma_{\text{eff}}(H)$ or $\gamma_{\text{eff}}(R)$ if one takes into account the following relationships: H (nm) = 0.3 S (nm), R (nm) = 0.3 S (nm) where H and R are the thickness and radius, correspondingly, of slit-like and cylindrical pores filled by water molecules with a diameter $d_0 \approx 0.3$ nm. As for the critical exponent α_{eff} , an essential decrease of the critical exponent γ_{eff} in a $3D \Leftrightarrow 2D$ dimensional crossover from $\gamma_{2D} = 1.75$ to $\gamma_{3D} = 1.25$ takes place within the same interval of H or R , which equals 0.9–3.6 nm because $\gamma_{\text{eff}}(0.9 \text{ nm}) = 1.63$ and $\gamma_{\text{eff}}(3.6 \text{ nm}) = 1.28$ (see Fig. 4).

3.2. Isothermal compressibility β_T

To study the influence of a $3D \Leftrightarrow 2D$ dimensional crossover on the isothermal compressibility β_T , it is necessary to use the following results:

1) in $3D$ case, the isothermal compressibility β_T in the bulk phase has a “strong” singularity at the critical temperature $T_c(\infty)$:

$$\beta_T^{3D} = \beta_{T_0}^{3D} \tau^{-\gamma_{3D}}, \quad (14)$$

where $\beta_{T_0}^{3D}$ is the non-universal amplitude of the isothermal compressibility, γ is the critical exponent, which equals $\gamma_{3D} = 1.250$ for the $3D$ Ising model;

2) in the $2D$ Ising model, the isothermal compressibility of a bulk fluid has a stronger divergence:

$$\beta_T^{2D} = \beta_{T_0}^{2D} \tau^{-\gamma_{2D}}, \quad (15)$$

where the critical exponent $\gamma_{2D} = 1.750$;

3) for fluids in a confined geometry, it is necessary to substitute the new temperature variable $\tau(S, \xi)$ given in Eq. (4) in (14) and (15) instead of the temperature variable $\tau = [T - T_c(\infty)]/T_c(\infty)$ for bulk fluids; 4) then the expressions of the isothermal compressibility in $3D$ and $2D$ confined fluids can be written in the following form:

$$\begin{aligned} \beta_T^{3D} &= \beta_{T_0}^{3D} \left\{ \left(\frac{G}{S} \right)^{\frac{1}{\nu}} + \right. \\ &+ \left. \left[1 + \left(\frac{G}{S} \right)^{\frac{1}{\nu}} \right] \left(\frac{\xi_0}{\xi} \right)^{\frac{1}{\nu}} \text{sign} \tau \right\}^{-\gamma_{3D}(S)}, \quad (16) \\ \beta_T^{2D} &= \beta_{T_0}^{2D} \left\{ \left(\frac{G}{S} \right)^{\frac{1}{\nu}} + \right. \end{aligned}$$

$$\left. + \left[1 + \left(\frac{G}{S} \right)^{\frac{1}{\nu}} \right] \left(\frac{\xi_0}{\xi} \right)^{\frac{1}{\nu}} \text{sign} \tau \right\}^{-\gamma_{2D}(S)}. \quad (17)$$

As in the previous case of the isochoric heat capacity C_V , the dependence of the isothermal compressibility β_T on the linear pore size S (thickness H or radius R) is given in accordance with the formula

$$\beta_T^* = \beta_T(S)/\beta_{T_0} = (S/G)^{\gamma_{\text{eff}}(S)/\nu_{\text{eff}}(S)}. \quad (18)$$

Equation (18) being quite similar to (12) contains the effective critical exponents $\gamma_{\text{eff}}(S)$ and $\nu_{\text{eff}}(S)$. Such an approach to describe a $3D \Leftrightarrow 2D$ dimensional crossover of the isothermal compressibility β_T^* gives the results presented in Fig. 5.

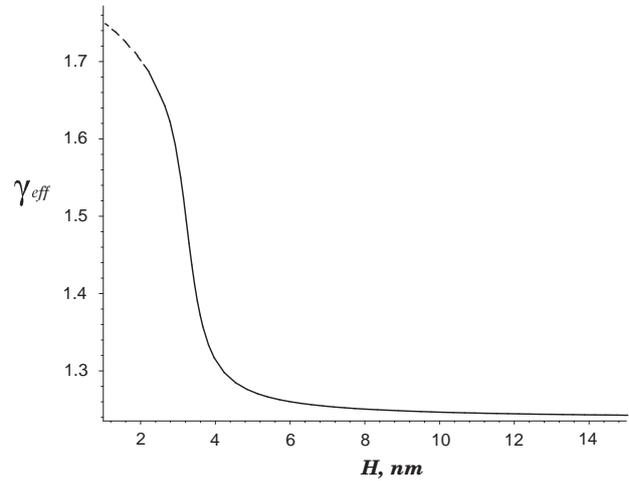


Fig. 4. Critical exponent γ_{eff} in a slit-like pore with thickness H filled by water

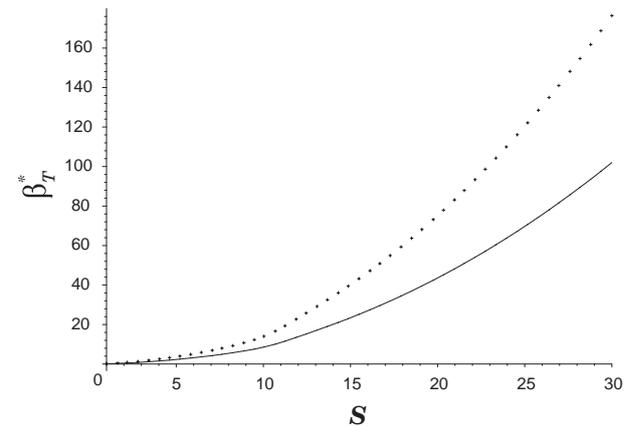


Fig. 5. Dependence of the isothermal compressibility β_T^* on S

The upper curve in Fig. 5 corresponds to cylindrical pores with $D_{\text{cros}} = 1$ and the geometrical factor $G = 2.40$, while the lower curve – to slit-like pores with $D_{\text{cros}} = 2$ and the geometrical factor $G = 3.14$.

4. Conclusion

In this paper, we have investigated the specific features of fluids in a reduced geometry. It is shown that, by considering the actual factor of a physical experiment such as the spatial limitation of fluid volumes under consideration, the critical exponents and thermophysical properties (namely, the heat capacity and the isothermal compressibility) may be essentially changed as compared with its critical behavior in bulk fluids. However, considering a factor of the spatial limitation and a lower crossover dimensionality successively into account enables us to support the hypothesis of the universality classes for confined fluids. We hope for that the further development of a consistent theory of the dimensional crossover in various phase transitions and critical phenomena will help one to stimulate new experiments in this difficult and important direction.

The authors wish to give their heartiest gratitude to Professor Leonid Bulavin for the friendship and the collaboration for many years and the fruitful guidance and advices, which are impossible to overestimate.

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Received 05.06.15

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РОЗМІРНИЙ КРОСОВЕР ТА
 ТЕРМОФІЗИЧНІ ВЛАСТИВОСТІ
 НАНОМАСШТАБНИХ КОНДЕНСОВАНИХ
 МАТЕРІАЛІВ

Резюме

Проблема, яка буде обговорюватися, полягає в такому: як результати для 3-вимірних систем переходять у результати для 2-вимірних систем і навпаки. Очевидно, що такий $3D \Leftrightarrow 2D$ розмірний кросовер має бути плавним та без порушень неперервності. Далі ця проблема вивчається для однокомпонентних класичних рідин в обмеженій геометрії, зокрема для плоско-паралельних і циліндричних пор з нижньою кросоверною розмірністю, відповідно, $D_{\text{cros}} = 2$ і $D_{\text{cros}} = 1$, які наповнені молекулами води. Вивчається вплив $3D \Leftrightarrow 2D$ розмірного кросовера на ефективні критичні індекси α_{eff} , ν_{eff} , γ_{eff} і на такі термофізичні властивості, як теплоємність C_V та ізотермічна стисливість β_T .