Scaling Migdal model and a nonparametric equation of state for argon

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Abstract. In this work, the nonparametric equation of state (NEOS) for matter is studied. We consider a methodical approach that is connected with some correlation between NEOS and Migdal’s scaling equation of state. We have analyzed the NEOS structure and have used additional functions depending on physical arguments (the density \( \rho \), the temperature \( T \)). Additionally, we have used a methodical approach connected with correlation between NEOS and Benedek’s hypothesis. A variant of NEOS, based on experimental data for argon has been developed. This input array includes \( C_V \) values, densities on the saturation line and data \((p, \rho, T)\). We have defined NEOS field of use: by density \( 0.67 \leq \rho/\rho_c \leq 1.22 \) at \( \rho < \rho_c \) and \( 0.97 \leq T/T_c \leq 1.08 \) at \( \rho > \rho_c \). Here \( \rho_c, T_c \) are critical parameters. NEOS is compared with experimental data on properties mentioned. These results are analyzed.

1. Introduction
An important physical problem is modeling of the thermodynamic surface in the region of strong density fluctuations. This problem has been investigated by many researchers [1–15]. There are interesting results obtained in [14, 15] and related to functions \( \rho^+(T), \rho^-(T) \) of metals (Al, Cu, U, . . . ) in the critical region, here \( \rho^+ \) is liquid density on saturation line, \( \rho^- \) is gas density, \( T \) is temperature. This problem is also considered in physics of high energy densities, e.g., one can find interesting \((p, \rho, T)\)-data which are obtained in [16–20] and related to some metals at high temperatures and high pressures including the critical region, where \( p \) is pressure.

The greatest attention is paid to the structure of the nonparametric equation of state (NEOS) where physical variables (the density \( \rho \) and the temperature \( T \)) are used [3–6]. Two directions of research exist. The first one is connected with some NEOS that meet certain requirements of the scaling theory of critical phenomena (ST). The authors of [7–10] have demonstrated some interesting results related to this direction.

The second direction includes NEOS with a structure that has few fitting coefficients and includes special functions [11–13]. For example, the special function \( h(x) \) is used in NEOS named as the Widom equation [21]: \( \Delta \mu = \Delta \rho|\Delta \rho|^{\delta-1}h(x) \), where \( \Delta \mu \) is the excess chemical potential, \( \rho_c, T_c \) are critical parameters, \( \Delta \rho = \rho/\rho_c - 1 \), \( \tau = T/T_c - 1 \), \( x = \tau/|\Delta \rho|^{1/\beta} \) is the scaling variable, \( \beta \) and \( \delta \) are critical indices. The \( h(x) \) function is called the scaling function related to the excess chemical potential. These variants of NEOS have allowed us to describe properties (pressure \( p \), isochoric heat capacity \( C_V \), isothermal compressibility coefficient \( K_T \)) in
The asymptotic neighborhood of the critical point with acceptable accuracy. Some discrepancy is realized in our tests:

- experimental data \((p, \rho, T)\) have deviated greatly from NEOS \([12]\);
- similar deviations of data \((C_V, \rho, T)\) are also realized.

The fundamental equations of state by Wagner et al \((1999)\) [22], Stewart and Jacobsen \((1989)\), Kozlov et al \((1996)\) [24] do not even qualitatively convey the behavior of argon in the critical region, and the error in describing isochoric heat capacity \((\text{Voronel’ et al} [25, 26])\) in the asymptotic neighborhood of the critical point reaches 80% or more \((\text{see, for example}, [22])\). The purpose of this paper is just to correct these shortcomings of equations of state \([3–13, 22–24]\).

We have set the task: to develop NEOS using the methodical approach connected with the equation of state in the form of the scaling Migdal model \([27]\). It is not difficult to adjust this NEOS to experimental data of argon. This input array has to include: values of \(C_V\), density on the saturation line and data for \(p, \rho, T\) in a wide range of applicability.

### 2. Joint analysis of the Migdal model and the Benedek model

The Migdal model \((\text{MM}) [27]\) is connected with the excess chemical potential \(\Delta \mu\) and written in the form

\[
\Delta \mu K^{(\gamma + \beta) / \gamma} = \psi_1 m + \psi_2 m^3 + \psi_3 m^5 + \ldots, \quad m = (\Delta \rho) K^{\beta / \gamma},
\]

where, \(\psi_1, \psi_2\) and \(\psi_3\) are constants; \(\beta\) and \(\gamma\) are critical indices; \(\Delta \mu = (\rho_c / \rho_c)|\mu(\rho, T) - \mu_0(T)|\) is the excess chemical potential; \(\mu(\rho, T)\) is the chemical potential; \(\mu_0(T)\) is regular function; \(K_T\) is the isothermal compressibility coefficient; \(\rho_c\) is the pressure in the critical point; \(\Delta \rho = \rho / \rho_c - 1\).

In our approach, the Benedek model \([28]\) that is connected with \(K_T\) and described by a scaling power law \([1]\) in a vicinity of the critical point is involved:

\[
K_T = D|\Delta \rho|^{-\frac{\gamma}{\beta}}|x + x_1|^{-\gamma},
\]

where \(D\) and \(x_1\) are constants; \(x = \tau / |\Delta \rho|^{1/\beta}\) is the scaling variable.

We have inserted \(K_T\) \((2)\) in equation \((1)\); it has allowed us to obtain \(\Delta \mu\) and the scaling function \(h(x)\) in the form

\[
\Delta \mu = (\Delta \rho)|\Delta \rho|^{\beta - 1} h(x),
\]

\[
h(x) = R\left[(x + x_1)^\gamma + \varepsilon (x + x_1)^{-\gamma + 1}\right],
\]

where \(R = \psi_1 / D; \varepsilon = (\psi_3 / \psi_1)D^{\beta - 1}\).

Compare \((3)\) with the linear model \((\text{LM}) [29]\)

\[
\Delta \mu = u_0|\rho_c\theta|\left(1 - \theta^2\right),
\]

where \(u_0\) is the individual constant. The LM model is presented in polar coordinates \((r, \theta)\):

\[
\Delta \rho = kr^\beta \theta, \quad \tau = r(1 - b^2 \theta^2),
\]

where \(k = (b_0^2 - 1)^{\beta} / x_0; x_0 = -x\) is the mass variable on the saturation line; \(b_0^2 = (\gamma - 2\beta) / [\gamma(1 - 2\beta)]\) \([30]\); \(\delta\) is the critical index.

With the usage of equation \((3)\), we write \((5)\) in the form

\[
\Delta \mu = (\Delta \rho)|\Delta \rho|^{\beta - 1} h_{\text{lm}}(x),
\]

where \(h_{\text{lm}}(x)\) is the scale function. The scale function \(h_{\text{lm}}(x)\) is written in the form:

\[
h_{\text{lm}}(x) = \frac{u_0}{k} \frac{1 - \theta^2}{\left|k|\theta|\right|^{\delta - 1}}.
\]
Find the parameters of function (4) using the equalities
\[ h(x = -x_0) = 0, \]
\[ h_i(x \to \infty) = h_{\text{lim}}(\theta = 0), \quad h_i(x = 0) = h_{\text{lim}}(\theta = \pm 1/b). \] (9, 10)

As a result, we get:
\[ h(x) = \frac{u_0 x_0^\gamma}{k} \left[ (\varphi + \varphi_1)^\gamma + \varepsilon (\varphi + \varphi_1)^{\gamma-4\beta} \right], \] (11)

where \( \varphi = x/x_0, \varphi_1 = x_1/x_0, \varphi_1 = -\varphi \) is the line of pseudo-critical points, \( \varepsilon = -(\varphi_1 - 1)^4\beta \).

We find the parameter \( \varphi_1 \) from equation [1]
\[ (\varphi_1)^\gamma + \varepsilon (\varphi_1 - 1)^{\gamma-4\beta} = \frac{b_0^{\delta-3}}{(b_0^2 - 1)^{\gamma-1}}. \] (12)

This line, \( \varphi_1 = -\varphi \), is determined by the equalities [31]: \( \frac{\partial \mu}{\partial s} = 0 \) and \( \frac{\partial \rho}{\partial p} \), where \( s \) is the entropy.

Due to MM and this methodical approach, we have got NEOS that can be written as:
\[ \Delta \mu = (\Delta \rho) |\Delta \rho|^{\delta-1} \frac{u_0 x_0^\gamma}{k} \left[ (\varphi + \varphi_1)^\gamma + \varepsilon (\varphi + \varphi_1)^{\gamma-4\beta} \right]. \] (13)

Our analysis has shown that NEOS (6) follows to some conditions of ST [32] operates with physical arguments and can be used to describe thermodynamic properties in the critical region.

3. Approbation of NEOS and numerical data on some thermodynamic properties of argon

In order to test NEOS structure, we have taken several steps. First, the scaling function \( h(x) \) on the basis of experimental data [25, 26, 33–35] of argon is investigated. The choice of the substance is done due to the facts:

- reliable data \( \rho, \rho, T \) are available [33];
- accurate experimental data \( \rho^+ , \rho^- , T \) in the critical region are available [34];
- extensive experimental information on \( C_V \) [25, 26, 33–35] is available.

These data are also used in some tests which have been done for crossover equations of state in [2, 3].

Second, we have considered the thermal equation of state, which has been developed on the basis of the Widom equation [21]. We have put \( \mu(\rho, T) \) (12) in a thermodynamic relation \( \rho F = \int \mu d\rho \) and obtained the following expression for the Helmholtz free energy \( F \):
\[ \frac{\rho}{p_c} F(\rho, T) = |\Delta \rho|^{\delta+1} a(x) + \frac{\rho}{p_c} A(\tau) + \frac{\rho}{p_c} B(\tau), \] (14)

where \( A(\tau) \) and \( B(\tau) = \mu_0(\tau) \) are regular functions; \( a(x) \) is the scale function:
\[ a(x) = \frac{u_0 \beta}{k x_1} |\Delta \rho|^{\delta-1} \left[ \int \frac{t^{1-\alpha}}{t} \left( 1 - \frac{\tau}{t} \right)^{2^{\delta-1}} dt - \frac{1}{x_1^{4\beta}} \int t^{1-\alpha} \left( 1 - \frac{\tau}{t} \right)^{6^{\beta-1}} dt \right] \]
\[ = \frac{u_0 \beta}{k x_1} \left( x + x_1 \right)^{2-\alpha} \left[ \frac{1}{2 - \alpha} + S_1 \right] - \frac{(x_1 - x_0)^{4\beta}}{x_1^{4\beta}} \left( \frac{1}{2 - \alpha} + S_2 \right), \] (15)
where $t = \tau + x_1|\Delta \rho|^{1/\beta}$,

$$S_1 = \sum_{n=1}^{N} (-1)^n \frac{(2\beta - 1) \cdots (2\beta - n)}{n!(2 - \alpha - n)} \left( \frac{x}{x + x_1} \right)^n,$$

$$S_2 = \sum_{n=1}^{N} (-1)^n \frac{(6\beta - 1) \cdots (6\beta - n)}{n!(2 - \alpha - n)} \left( \frac{x}{x + x_1} \right)^n.$$  \hspace{1cm} (16)  

We put $F(\rho, T)$ (14) into a thermodynamic relation $p = \rho^2 (\partial F/\partial \rho)_T$ and obtain the thermal equation of state

$$\pi = \omega(\Delta \rho)|\Delta \rho|^{\delta-1} h(x) - |\Delta \rho|^{\delta+1} a(x) - \frac{\rho_c}{p_c} A(\tau),$$

where $A(\tau)$ is regular function of temperature; $\omega = \rho/\rho_c$; $\pi = p/p_c$.

Third, to represent $A(T)$, which is included in (18), the following form is chosen

$$\frac{\rho_c}{p_c} A(T) = 1 + \sum_{i=1}^{N_1} c_i \tau^i.$$  \hspace{1cm} (19)  

Then we put $F(\rho, T)$ (14) into a thermodynamic relation $C_V = -T(\partial^2 F/\partial T^2)_V$ and get $C_V$ in the form

$$\frac{\rho TC}{p_c T} C_V = -|\Delta \rho|^{\delta} \tilde{a''}(x) - \frac{\rho_c}{p_c} A''(T) - \omega \frac{\rho_c}{p_c} B''(T),$$

where

$$\frac{\rho_c}{p_c} A''(T) = \sum_{i=2}^{N_1} i(i-1)c_i \tau^{i-2},$$  \hspace{1cm} (21)  

$$\frac{\rho_c}{p_c} B''(T) = \sum_{i=2}^{N_2} i(i-1)d_i \tau^{i-2}.$$  \hspace{1cm} (22)  

Fourth, to represent $a''(x)$ or the second derivative of function $a(x)$, the following form has been obtained

$$a''(x) = \frac{u_0 \beta \gamma_1}{k x_1^{2\beta}} |\Delta \rho|^{\alpha/\beta} \left[ \int t^{-\alpha-1} \left( 1 - \frac{\tau}{t} \right)^{2\beta-1} \frac{dt}{x_1^{4\beta}} + \int t^{-\alpha-1} \left( 1 - \frac{\tau}{t} \right)^{2\beta-1} \frac{dt}{x_1^{4\beta}} \right]$$

$$= -\frac{u_0 \beta \gamma_1}{k x_1^{2\beta}} (x + x_1)^{-\alpha} \left[ \left( \frac{1}{\alpha + S_1''} \right) - \varepsilon_1 \frac{(x_1 - x_0)^{4\beta}}{x_1^{4\beta}} \left( \frac{1}{\alpha + S_2''} \right) \right],$$

where $\varepsilon_1 = (\gamma - 4\beta)(\gamma - 4\beta - 1)/\gamma_1$,

$$S_1'' = \sum_{n=1}^{N} (-1)^n \frac{(2\beta - 1) \cdots (2\beta - n)}{n!(\alpha + n)} \left( \frac{x}{x + x_1} \right)^n,$$

$$S_2'' = \sum_{n=1}^{N} (-1)^n \frac{(6\beta - 1) \cdots (6\beta - n)}{n!(\alpha + n)} \left( \frac{x}{x + x_1} \right)^n.$$  \hspace{1cm} (23)  

We have concluded that the exact solution of equations (14), (20) corresponds to functions $a(x)$ (15)–(17) and $a''(x)$ (23)–(25) at $N = \infty$. However, our analysis has shown that the error, $\delta X = (X^{(e)} - X^{(r)})/X^{(e)}100\%$, practically does not change if we use the second variant of $N$, namely $N = 10$, where $(e)$ is an index corresponding to experimental data [25, 26, 33–35], $(r)$ is an index corresponding to $X$ values calculated. We have considered the following properties:
Figure 1. Isotherms and the coexistence line of argon: 1—the coexistence line calculated with the use of equation (26); 2—experimental data \((\rho^\pm, T)\) from [34]; isotherms: 3, 4, 5, 6, 7—\(p, \rho, T\) values calculated with the use of equation (23); 8, 9, 10, 11, 12—experimental data \((p, \rho, T)\) from [33]; 3, 8—\(T = 163.15\) K; 4, 9—\(T = 158.15\) K; 5, 10—\(T = 153.15\) K; 6, 11—\(T = 151.65\) K; 7, 12—\(T = 150.65\) K.

- the value \(X = p\); the values \(p^{(r)}\) are obtained on the basis of equation of state (18);
- the value \(X = C_V\); the values \(C_V^{(r)}\) are calculated with the use of equation (20).

We have considered variants when \(N_1 = 2\) and \(N_2 = 3\). As a result, from equations (18), (20) we have obtained the following expressions

\[
\pi = 1 - c_1 \tau - c_2 \tau^2 + (\Delta \rho) |\Delta \rho|^{\delta-1} h(x) - |\Delta \rho|^{\delta+1} a(x),
\]

\[
\frac{\rho T^2}{p_c T} C_V = -2c_2 - 2\omega - 6\omega \tau - |\Delta \rho|^{\frac{2}{\delta}} a''(x). \tag{27}
\]

We can find the coefficients of equations (26), (27) by minimizing the functional \(U\); has the form

\[
U = \sum_{n=1}^{N_1} \left[ Q_{p,s,n} (p^{(r)}_{s,n} - p^{(e)}_{s,n}) \right]^2 + \sum_{n=1}^{N_2} \left[ Q_{p,n} (p^{(r)}_n - p^{(e)}_n) \right]^2 + \sum_{n=1}^{N_3} \left[ Q_{C_V,n} (C_V^{(r)} - C_V^{(e)}) \right]^2,
\]

where \(Q^*_{X,n}\) is the weight of point \(n\), \(X\) is the name of the property; \(N_1, N_2, N_3\) are the numbers of points related to experimental data \((p_s, \rho^\pm, T), p(\rho, T), C_V(\rho, T)\) [20, 21, 27–29].

As a result, the following values are assigned to the parameters and coefficients of (26), (27): \(c_1 = -6.084838 \times 10^5\), \(c_2 = 12.507385 \times 10^4\), \(d_2 = 5.303902 \times 10^4\), \(d_3 = 1.581781 \times 10^3\), \(u_0 = 14.881843 \times 10^3\), \(x_0 = 0.310661 \times 10^6\), \(\varphi_1 = 2.0925\), \(\beta = 0.325\), \(\gamma = 1.24\), \(N = 10\), \(N_1 = 2\), \(N_2 = 3\), \(T_c = 150.66\) K, \(p_c = 4.8634\) MPa, \(\rho_c = 535.1\) kg/m\(^3\). The remaining critical indices \((\alpha = 0.11; \delta = 4.815)\) are determined on the basis of Griffith equations: \(2 - \alpha = \beta \delta + \beta\) and \(\gamma = \beta \delta - \beta\) [30].

Properties \((p, \rho^-, \rho^+, C_V)\) have been calculated from equations (26), (27) in the region of highly developed density fluctuations. Our comparisons have allowed us to estimate that these calculated values coincide with the experimental data mainly within the limits of experimental errors (figures 1–3).
Figure 2. The heat capacity $C_V$ in the single phase region: 1, 3, 5, 7, 9, 11—experimental data $(C_V, \rho, T)$ from [25,26]; the isochores: 1—$\rho = 504 \text{ kg/m}^3$; 3—$\rho = 533 \text{ kg/m}^3$; 5—$\rho = 535 \text{ kg/m}^3$; 7—$\rho = 538 \text{ kg/m}^3$; 9—$\rho = 549 \text{ kg/m}^3$; 11—$\rho = 560 \text{ kg/m}^3$; 13—$\rho = 666 \text{ kg/m}^3$; 2, 4, 6, 8, 10, 12—data $(C_V, \rho, T)$ calculated with the use of equation (27).

Figure 3. Behavior of $C_V$ in the single phase region: 1—experimental data $(C_V, \rho, T)$ from [34], $\rho = 565.5 \text{ kg/m}^3$; 2—experimental data $(C_V, \rho, T)$ from [26], $\rho = 560 \text{ kg/m}^3$; 3—data $(C_V, \rho, T)$ calculated with the use of equation (27), $\rho = 565.5 \text{ kg/m}^3$; 4—data $(C_V, \rho, T)$ calculated with the use of equation [22], $\rho = 565.5 \text{ kg/m}^3$.

We can conclude:

- nonparametric equation of state (14) and scaling function (15) are physically justified;
- in the critical region, this equation of state transfers quantitatively the thermal surface just like the caloric surface.
4. Discussion
We have studied the scaling Migdal model [27] and the Benedek hypothesis [28]. Our approach allows us to explain the results obtained in [1].

One can see a good correlation of calculated properties \((p, \rho^-, \rho^+)\) on the saturation line with corresponding experimental values. There exists good agreement between the experimental \(p(\rho, T)\) function and corresponding data calculated with the use of equation (26). Just the same situation is realized for the experimental \(C_V(\rho, T)\) function and corresponding data calculated with the use of equation (27).

Our analysis has shown (see figures 1–3) that the working region of the equation of state (14) has the following borders:

- they are \(0.67 \leq \rho/\rho_c \leq 1.22\) in density;
- they are \(0.99 \leq T/T_c \leq 1.08\) in temperatures if \(\rho < \rho_c\);
- they are \(0.997 \leq T/T_c \leq 1.08\) if \(\rho > \rho_c\).

5. Conclusion
Thus, the working region of the equation of state proposed in this paper is not inferior to the working area of LM and the cubic Litster–Ho model. It is also shown that the equation of state of this paper faithfully conveys the thermal surface outside the working area. It is possible to conclude that the equation of state of this paper can be used in constructing both crossover equations of state, and large-scale combined equations of state satisfying ST.

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