

EQUATION OF STATE AND EQUATIONS OF THERMODYNAMIC PROCESSES IN DENSE LIQUIDS AND GASES (NITROGEN AND ARGON)

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An isothermal equation of state of dense liquids and gases is suggested that describes, with rather high accuracy, the available experimental data for nitrogen and argon within a wide range of temperatures and allows one to perform extensive extrapolation with respect to density.

In a great number of engineering devices nitrogen and argon are used as a working body. To design these devices and technological processes implemented by them one should know the equation of state of liquefied and gaseous nitrogen and argon at high pressures within a wide range of temperatures.

A great number of the equations of state are known that describe the thermodynamic properties of nitrogen and argon dense liquids and gases [1-7]. However, only some of them allow the determination of parameters within the range of pressures in which experimental data are absent. These include various modifications of the Tate equation [2]. At $T = \text{idem}$ this equation has the form

$$v = v_0 [1 - C \ln (p + B)/(p + B_0)], \quad (1)$$

where $B = B(T)$; $C = 0.3678$. It properly describes the volumetric behavior of nitrogen and argon at pressures of from 300 to 1200 MPa. The Tate equation of state is among those that have been theoretically substantiated [8, 9]. However, it has the crucial drawback that at a very high pressure volume becomes negative. A simple empirical equation of state suggested in [7] is free of this drawback

$$\sqrt{\rho} = a + b \ln p, \quad (2)$$

where $a = a(T)$; $b = b(T)$. But this equation is unsuitable for determining the volume of substance in a metastable state (a superheated liquid) at negative absolute pressures [10].

In [11, 12] it is shown that the equation of an isentropic process in a liquid has the form

$$v = v_0 [(p_0 + B_s)/(p + B_s)]^{1/\kappa} \quad \text{or} \quad \rho = \rho_0 [(p + B_s)/(p_0 + B_s)]^{1/\kappa}, \quad (3)$$

where $B_s = B_s(s)$; $\kappa = \kappa(s)$.

Calculations showed that the isothermal equation of state

$$v = v_0 [(p_0 + B)/(p + B)]^{1/n} \quad \text{or} \quad \rho = \rho_0 [(p + B)/(p_0 + B)]^{1/n}, \quad (4)$$

where $B = B(T)$; $n = n(T)$, which is analogous to expression (3), holds for many liquids and dense gases if $\rho/\rho_{cr} \geq \rho_{\min}/\rho_{cr}$.

Table 1 gives the values of the parameters n and B of Eq. (4) for known experimental isotherms of nitrogen dense liquid and gas (for $\rho/\rho_{cr} \geq \rho_{\min}/\rho_{cr} = 2$). Here the relative mean and maximum deviations (δ) of the values of nitrogen density calculated by equation of state (4) from the experimental data on each isotherm are presented. The table also gives similar deviations for Eq. (2). It follows from the table that the deviation of the results of

TABLE 1. Values of the Coefficients of the Equation of State (4) for Nitrogen and Relative Deviations of the Values of Molar Volumes of Nitrogen Calculated by Eqs. (2) and (4) from the Test Data on Experimental Isotherms

Isotherm, K	Maximum pressure on isotherm, MPa	Reference	Coefficients of Eq. (4)		δ , % (numerator – mean deviations; denominator – maximum deviations)	
			n	B , MPa	Eq. (2)	Eq. (4)
115.66	295	[2, p. 112-113]	6.6711	11.46	0.02/0.04	0.03/0.06
145.08	532.5		6.5068	-2.59	0.06/0.19	0.4/0.72
175.83	834.6		5.5184	-15.88	0.10/0.20	0.06/0.10
205.52	998.8		5.1865	-25.74	0.08/0.13	0.03/0.07
252.56	1055.4		4.3334	-39.13	0.04/0.10	0.02/0.07
280.93	1056.7		4.6310	-44.23	0.06/0.15	0.05/0.19
308.79	1051.2		4.4641	-54.21	0.09/0.12	0.021/0.30
247.5	1600	[2, p. 115]	4.7231	-41.65	0.12/0.22	0.03/0.08
273.3	2000		4.5212	-45.21	0.15/0.60	0.03/0.09
297.4	2200		4.4452	-50.77	0.15/0.62	0.02/0.05
320.8	2200		4.3599	-56.55	0.15/0.41	0.02/0.04
294.65	658.6	[13]	4.4138	-51.20	0.13/0.21	0.05/0.14
453.15	506.6		4.1500	-103.67	0.09/0.16	0.02/0.04
473.15	810.6		4.0527	-105.68	0.30/0.73	0.04/0.07
573.15	1013.2		3.8279	-117.81	0.37/1.14	0.16/0.36
673.15	1013.2		3.7409	-126.63	0.46/1.00	0.20/0.37
308.15	1000	[2, p. 109]	4.4301	-51.63	0.05/0.32	0.03/0.08
373.15	1000		4.1393	-63.27	0.15/0.47	0.07/0.22
473.15	1000		4.1012	-110.09	0.25/0.79	0.08/0.17
573.15	1000		3.8165	-115.90	0.19/0.68	0.06/0.13
673.15	1000		3.7510	-125.71	0.23/0.61	0.11/0.23

calculations by Eq. (4) from the test data does not exceed the error of the experiment and that Eq. (4) gives better results than does Eq. (2).

In Tables 2 and 3 for nitrogen and argon dense liquids and gases values of the parameters n and B and the derivatives dn/dT and dB/dT are given that were obtained from the experimental values presented in [2] and from tables in [1, 5].

In Fig. 1 the curves $n = n(\tau)$ and $B/\rho_{cr}(\tau)$ for nitrogen and argon are presented. Both curves are virtually the same, i.e., nitrogen and argon dense liquids and gases at $0.6 < \tau < 3$ are thermodynamically similar in their isothermal processes.

Equation (4) holds for argon, if $\rho/\rho_{cr} \geq \rho_{min}/\rho_{cr} = f(\tau)$ (see Fig. 1). It is seen from the figure that the parameters n and B are the monotonically decreasing functions of τ . They are independent of the value of initial pressure p_0 if $\rho/\rho_{cr} \geq \rho_{min}/\rho_{cr}$. Near the critical temperature ($\tau \approx 1$) the parameter B changes its sign: in liquid B is positive and in a dense gas it is negative. The parameter B , which has the dimension of pressure, is associated with the value of internal pressure:

$$p_{in} = (\partial u / \partial v)_T = T (\partial p / \partial T)_v - p = (\gamma / \gamma_0 - 1) = (\alpha / \alpha_0) n (p + B) - p.$$

TABLE 2. Values of the Parameters n , B , dn/dT , dB/dT for Nitrogen Dense Liquid and Gas

T , K	n	B , MPa	dn/dT , 1/K	dB/dT , MPa/K
65	13.5	35.9	—	—
80	8.85	31.9	-0.084	-0.62
100	7.70	15.5	-0.050	-0.77
125	6.75	2.0	-0.033	-0.46
150	6.03	-8.2	-0.024	-0.41
200	5.22	-26.5	-0.013	-0.33
250	4.70	-42	-0.08	-0.29
300	4.45	-54	-0.06	-0.27
350	4.30	-67	-0.05	-0.255
400	4.18	-81.5	-0.04	-0.24
450	4.10	-94	-0.03	-0.22
500	4.00	-104	-0.02	-0.20
550	3.93	-112	-0.017	-0.18
600	3.85	-118	-0.017	-0.17
650	3.8	-126	-0.017	-0.17
700	3.7	-137	-0.017	-0.17
750	3.6	-146	-0.017	-0.17
800	3.5	-155	-0.017	-0.17

TABLE 3. Values of the Parameters n , B , dn/dT , dB/dT for Argon Dense Liquid and Gas

T , K	n	B , MPa	dn/dT , 1/K	dB/dT , MPa/K
90	9.5	44	-0.09	-0.8
100	8.52	37	-0.082	-0.7
110	7.90	30	-0.03	-0.75
120	7.70	20	-0.015	-0.79
130	7.55	12	-0.015	-0.80
140	7.35	5	-0.015	-0.78
150	7.12	-1.5	-0.025	-0.73
160	6.75	-6.5	-0.032	-0.60
170	6.40	-10	-0.032	-0.40
180	6.10	-13	-0.032	-0.30
190	5.80	-15.5	-0.032	-0.25
200	5.50	-18	-0.03	-0.25
210	5.35	-21	-0.026	-0.25
220	5.25	-24	-0.020	-0.34
230	5.15	-28	-0.015	-0.40
240	5.05	-32	0.010	-0.47
250	5.00	-35	0.009	0.45
300	4.80	-59	0.005	0.38
400	4.45	-92	0.004	0.27
500	4.15	-113	0.003	0.17
600	3.85	-123	0.0025	0.09
673	3.65	-127	0.002	0.05

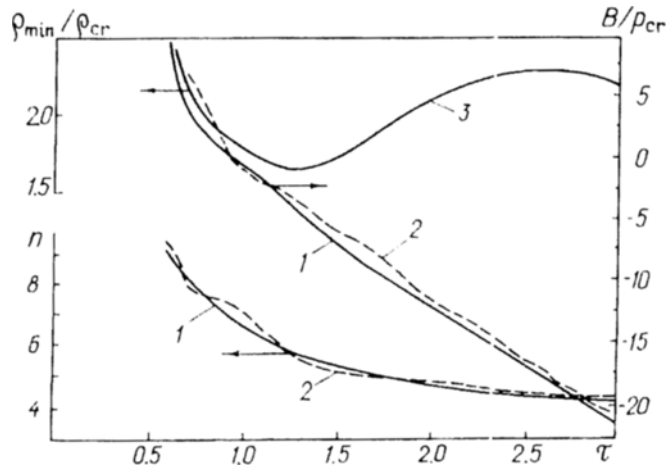


Fig. 1. The values of the parameters n and B/ρ_{cr} from Eq. (4) for nitrogen and argon dense liquids and gases, which holds at $\rho/\rho_{cr} \geq 2$ for nitrogen and $\rho/\rho_{cr} \geq \rho_{min}/\rho_{cr}$ for argon: 1) nitrogen, 2) argon, 3) ρ_{min}/ρ_{cr} for argon.

With the quantities n , B , dn/dT , and dB/dT being known, the isothermal entropy difference is found from the equation

$$\begin{aligned}
 s - s_0 = & \int_{p_0}^p (\partial v / \partial T)_p dp = - (\partial v_0 / \partial T)_p (p_0 + B) \frac{n}{n-1} \left\{ \left[(p+B)/(p_0+B) \right]^{\frac{n-1}{n}} - 1 \right\} - \\
 & - \frac{v_0 (p_0 + B)}{n(n-1)} \frac{dn}{dT} \left\{ \left(\frac{p+B}{p_0+B} \right)^{(n-1)/n} \ln \left(\frac{p+B}{p_0+B} \right) - \frac{n}{n-1} \left[\left((p+B)/(p_0+B) \right)^{\frac{n-1}{n}} - 1 \right] \right\} + \\
 & + v_0 dB/dT \left\{ \frac{p-p_0}{p_0+B} \left[(p_0+B)/(p+B) \right]^{1/n} - \frac{n}{n-1} \left[\left((p+B)/(p_0+B) \right)^{\frac{n-1}{n}} - 1 \right] \right\}. \quad (5)
 \end{aligned}$$

The isothermal enthalpy difference is determined by the expression

$$h - h_0 = \int_{p_0}^p [v - T (\partial v / \partial T)_p] dp = (s - s_0) T + \frac{v_0 (p_0 + B) n}{n-1} \left[\left(\frac{p+B}{p_0+B} \right)^{\frac{n-1}{n}} - 1 \right]. \quad (6)$$

The equation for volatility has the form

$$\ln (f/f_0) = \frac{1}{RT} \int_{p_0}^p v dp = \frac{v_0 (p_0 + B)}{RT} \frac{n}{n-1} \left\{ \left[(p+B)/(p_0+B) \right]^{\frac{n-1}{n}} - 1 \right\}.$$

To estimate the possibilities of extensive extrapolation with respect to pressure for argon, approximate values of n and B were found using the data of [2, pp. 117-118] at $T = 348.15$ K on the basis of the volume values at pressures of 245.2, 294.2, and 343.2 MPa. The maximum value of δ obtained in calculations by Eq. (4) with approximate n and B ($n = 4.34$, $B = 60.5$ MPa) at a pressure of 1667.1 MPa (i.e., 4.86 times higher than the maximum used in determination of n and B) is 1.48%.

Analysis of the results of calculations on the basis of tables of [1, 5] allows one to conclude that in nitrogen and argon dense liquids and gases thermodynamic processes at a constant value of some function of state ($z = T, s, h, \dots$) are described by the equation

$$(p + B_z) v^{nz} = \text{idem}, \quad (7)$$

where $B_z = B_z(z)$, $n_z = n_z(z)$.

Pressure and volume on the lines of solidification and melting (in liquid and solid phases), on the line of boiling (at $\rho/\rho_{cr} \geq \rho_{min}/\rho_{cr}$), and also for argon in a crystalline state at constant z (from the data of Table VIII from [5]) are related by the same equations.

The line of melting of solid argon at $T = 86-107$ K is described, with a maximum error of 0.1%, by the equation

$$v_T = v_{T0} [(p_0 + B'_\lambda)/(p + B'_\lambda)]^{1/n'_\lambda}, \quad (8)$$

where $n'_\lambda = 3.8$, $B'_\lambda = 861.67$ MPa (by the data of [5]).

Specific volumes of liquefied argon on the solidification line up to $T = 300$ K were found with the aid of Eq. (4) and Table 3. Pressure of solidification and melting is determined by formula (1.68) from [5]:

$$p = 0.499313 (T - 30.179)^{1.43057} - 148.494 \text{ MPa}, \quad (9)$$

which holds up to $T = 273$ K. For $T > 273$ K the solidification pressure was found from the verified formula (9) for obtaining an agreement with the experimental value of the solidification pressure ($p_{sol} = 1324$ MPa at $t = 22^\circ\text{C}$ (Table 5.12, [2])):

$$p = 0.499313 (T - 30.179)^{1.43057} - 148.494 - 0.4818 (T - 273.15) \text{ MPa}.$$

Specific volumes of liquefied argon on the line of solidification at $T = 88-300$ K are found, with a maximum error of 0.8%, by the equation

$$v = v_0 [(p_0 + B''_\lambda)/(p + B''_\lambda)], \quad (10)$$

where $n''_\lambda = 6.21$, $B''_\lambda = 243.695$ MPa.

Specific volumes of liquefied argon on the boiling line at $\rho/\rho_{cr} \geq 2.12$ ($T \leq 123$ K) are found, with a maximum error of 0.8%, by the equation

$$v = v_0 [(p_0 + B'_\lambda)/(p + B'_\lambda)]^{1/n'_\lambda}, \quad (11)$$

where $n'_\lambda = -7.32$, $B'_\lambda = 0.29456$ MPa.

Specific volumes of liquefied nitrogen on the solidification line up to $T = 300$ K were found by Eq. 4 and Table 2. The pressure on the line of solidification is determined by the Mills and Grilli equation [1], which holds up to 300 K:

$$p_{sol} = -160.662 + 0.0957894 T^{1.791} \text{ MPa}.$$

Within the entire range of temperatures of from 65 to 300 K specific volumes of nitrogen are determined, with a maximum error of 0.016%, by Eq. (10), where $n''_\lambda = 4.587$, $B''_\lambda = 384.064$ MPa.

Specific volumes of liquefied nitrogen on the line of boiling for $T \leq 100$ K are described, with a maximum error of 0.87%, by Eq. (11), where $n'_\lambda = -7.78$, $B'_\lambda = 0.15131$ MPa.

Just as in Eq. (4), in Eqs. (7), (8), (10), and (11) the values of the parameters n_z and B_z do not depend on the value of p_0 , if $\rho_0/\rho_{cr} \geq \rho_{min}/\rho_{cr}$.

In Eq. (7) the parameters n_z and B_z are determined by the following relations

$$n_z = \frac{1/\beta_z - 1/\beta_{z0}}{p - p_0} = \frac{m_z p - m_{z0} p_0}{p - p_0} = m_z + p (\partial m_z / \partial p)_z = \frac{K_z - K_{z0}}{p - p_0} = (\partial K_z / \partial p)_z = n_z(z);$$

$$B_z = (m_z - n_z) p / n_z = 1/(\beta_z n_z) - p = B_z(z). \quad (12)$$

In dense liquids and gases the Grüneisen parameter [12]

$$\theta = \frac{\rho}{T} \left(\frac{\partial T}{\partial \rho} \right)_s = v \left(\frac{\partial p}{\partial u} \right)_v = \frac{\alpha a^2}{c_p} = \frac{\alpha v}{\beta c_v} = \frac{c_p/c_v - 1}{\alpha T} \quad (13)$$

is a slowly varying quantity. If at $s = \text{idem}$ it is assumed to be constant, then we obtain the equation for an isentrope in the form

$$T/T_0 = [(p + B_s)/(p_0 + B_s)]^{\theta/\kappa} = (\rho/\rho_0)^\theta. \quad (14)$$

Specific work of compression and expansion at $s = \text{idem}$ and $\kappa = \text{idem}$ is determined by the formulas [12]

$$L_{s\text{com}} = h_{2s} - h_1 = \int_1^{2s} dp/\rho = \frac{\kappa (p_1 + B)}{(\kappa - 1) \rho_1} \left[\left(\frac{p_2 + B}{p_1 + B} \right)^{\frac{\kappa-1}{\kappa}} - 1 \right]; \quad (15)$$

$$L_{s\text{exp}} = h_1 - h_{2s} = \int_{2s}^1 dp/\rho = \frac{\kappa (p_1 + B)}{(\kappa - 1) \rho_1} \left[1 - \left(\frac{p_2 + B}{p_1 + B} \right)^{\frac{\kappa-1}{\kappa}} \right].$$

The following examples allow one to estimate the accuracy of the suggested equation of state and the equations of thermodynamic processes for nitrogen and argondense liquids and gases.

Example 1. To determine thermodynamic parameters of nitrogen at $T = 100$ K and $p = 100$ MPa.

From the tables of [1] at $T = 100$ K and $p = 1.0$ MPa we take the known values: $\rho = 689.64$ kg/m³, $s_0 = 3.375$ kJ/(kg·K), $h_0 = 174.6$ kJ/kg, $dv_0/dT = 1.2862 \cdot 10^{-5}$ m³/(kg·K). From Table 2 we find $n = 7.70$, $B = 15.5$ MPa, $dn/dT = -0.048$ 1/K, $dB/dT = -0.76$ MPa/K. From Eqs. (4)-(6) at $p = 100$ MPa we obtain the following values: $\rho = 887.91$ kg/m³, $s = 2.890$ kJ/(kg·K), $h = 247.9$ J/kg. The errors of the determination of the parameters, as compared with [1], are 0.036% for density, 0.01% for entropy, and 0.04% for enthalpy.

Example 2. To determine thermodynamic parameters of nitrogen after isentropic compression to $p = 39$ MPa and $T = 150$ K. The initial parameters: $T = 110$ K, $p_0 = 1.5$ MPa, $s_0 = 3.600$ kJ/(kg·K), $\rho_0 = 621.53$ kg/m³, $h = 199.0$ kJ/kg.

Using the values of pressure and density at three points from [1] and the equations

$$\frac{p_2 - p_0}{p_2 - p_1} \left(\frac{\rho_1}{\rho_0} \right)^{n_z} - \frac{p_1 - p_0}{p_2 - p_1} \left(\frac{\rho_2}{\rho_0} \right)^{n_z} = 1; \quad B_z = \frac{p_2 - p_0 (\rho_2/\rho_0)^{n_z}}{(\rho_2/\rho_0)^{n_z} - 1} \quad (16)$$

and Eq. (13) at $z = s = 3.600$ kJ/(kg·K), we find $n_z = \kappa = 8.69$, $B_s = 13.9126$ MPa, $\theta = 1.4168$. At $p = 39$ MPa from Eqs. (3), (14), and (15) we obtain: $\rho = 716.3$ kg/m³, $T = 134.49$ K, $h = 254.45$ kJ/kg. The errors of determination of density are 0.76%, enthalpy 0.13%, and temperature 1.89%.

At the same values of κ , B_s , and θ and finite $T = 150$ K from Eqs. (14) and (15) we obtain $p = 89.373$ MPa, $\rho = 773.62$ kg/m³, $h = 321.8$ kJ/kg. The errors of determination of pressure are 0.003%, density 0.002%, and enthalpy 0.31%.

Example 3. To determine nitrogen density after adiabatic throttling at $h = 230.5$ kJ/kg, to $p = 34.375$ and 57.155 MPa. The initial parameters: $p_0 = 100$ MPa, $T_0 = 90$ K, $\rho_0 = 910.47$ kg/m³, $s = 2.706$ kJ/(kg·K).

Using the values of pressure and density at three points from [1], we find by Eqs. (16) at $z = h = 230.5$ kJ/kg = idem $n_h = 3.85$; $B_h = 16.52$ MPa. By Eq. (7) at $p = 34.375$ MPa we find $\rho = 734.23$ kg/m³; at $p = 57.155$ MPa we have $\rho = 808.27$ kg/m³. The error of density determination in the both cases is 0.21%.

Example 4. To determine the thermodynamic properties of argon at $T = 100$ K in a stable state at $p = 30$ MPa and in a metastable state at $p = -10$ MPa.

We assume the thermodynamic parameters of argon from [5] at $T = 100$ K and $p = 1$ MPa to be known: $v_0 = 0.7611 \cdot 10^{-3}$ m³/kg, $h_0 = 90.08$ kJ/kg, $s_0 = 1.527$ kJ/(kg·K), $(\partial v_0/\partial T)_{\text{exp}} = 3.96 \cdot 10^{-6}$ m³/(kg·K). From Table

3 we have $n = 8.52$, $B = 37$ MPa, $dn/dT = -0.082$ 1/K, $dB/dT = -0.7$ MPa/K. By Eqs. (4)-(6) we obtain at $p = 30$ MPa: $v = 0.7121 \cdot 10^{-3}$ m³/kg, $h = 102.20$ kJ/kg, $s = 1.435$ kJ/(kg·K). The errors of determination of the specific volume are 0.013%, enthalpy 0.25%, and entropy 0.20%.

At $p = -10$ MPa from Eqs. (4)-(6) we obtain $v = 0.7923 \cdot 10^{-3}$ m³/kg, $h = 86.83$ kJ/kg, $s - s_0 = 0.0528$ kJ/(kg·K). In Table 5.6 of [10] the following values are given: $v = 0.795 \cdot 10^{-3}$ m³/kg, $h = 86.65$ kJ/kg, $s - s_0 = 0.053$ kJ/(kg·K). The errors of determination of the specific volume are 0.34%, enthalpy 0.21%, and the difference $s - s_0$ 0.4%.

Example 5. To determine temperature in crystalline argon after omnidirectional shock (isentropic) compression to $p = 50$ MPa. The initial parameters: $s = 0.8250$ kJ/(kg·K), $T_0 = 70$ K, $p = 0.1$ MPa, $v_0 = 0.6008 \cdot 10^{-3}$ m³/kg. Using the values of pressure and specific volume of argon at three points from [5], we obtain by Eqs. (16) and (13) at $z = s = 0.8250$ kJ/(kg·K) the values of $\kappa = 7.4$, $B_s = 275.4$ MPa, $\theta = 2.799$. From Eq. (14) we find $T = 74.55$ K. The error of temperature determination is 0.02%.

Thus, the suggested equation of state and the equations of thermodynamic processes (7) can be recommended for practical calculations of thermodynamic parameters of nitrogen and argon dense liquids and gases at high pressures within a wide range of temperatures, including extensive extrapolation of the known data to higher pressures.

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NOTATION

v , specific volume; p , pressure; p_0 , pressure taken as zero for reference; v_0 , specific volume at this pressure; ρ , density; s , entropy; T , absolute temperature; h , enthalpy; f , volatility; $\delta = |v_{\text{exper}} - v_{\text{calc}}|/v_{\text{calc}}$, %, relative deviation; R , universal gas constant; u , internal energy; $\gamma/\gamma_0 = T(\partial p/\partial T)_v/p$, thermal coefficient of pressure; $\alpha/\alpha_0 = (T/v)(\partial v/\partial T)_{\text{exp}}$, relative coefficient of volumetric expansion; $\beta_z = -(\partial v/\partial p)_z/v$, coefficient of compressibility in the process of $z = \text{idem}$; $m_z = \rho(\partial p/\partial \rho)_z/p = 1/(\beta_z p)$; $K_z = v(\partial p/\partial v)_z$, elasticity modulus in the process of $z = \text{idem}$ [14]; a , sound velocity; c_p , heat capacity at constant pressure; c_v , heat capacity at constant volume; $\tau = T/T_{\text{cr}}$. Subscript cr refers to critical parameters.

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