

A few-parameter equation of state of the condensed matter

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Abstract. A simple caloric equation-of-state model is proposed to describe thermodynamic properties of solid materials without phase transitions with the minimum number of parameters as initial data. The thermal vibrations of the crystal lattice are described by the Debye approximation. The parameter values on the zero isotherm are calculated analytically from the generalized form of the Grüneisen function. Thermodynamic characteristics are calculated in the wide range of densities and pressures. The results of the theoretical calculations for these materials are exhaustively compared with the available experimental data for high energy densities.

1. Introduction

The modern wide-range equations of state, constructed to describe the behavior of metals in a wide range of compression parameters, contain dozens of free parameters and experimentally found constants [1, 2]. Sometimes, these models are complemented by taking into account multiple solid phases [3], the number of constants in this case inexorably increases. The constants are calculated according to the shock-wave experiments, determined from isentropic curves of unloading of porous samples, or are found from the experimental data in a wide area of the phase diagram from thermodynamic relations. It is worth mentioning the modern models with fewer constants as the initial data [4, 5], but still there are too many of them to solve real engineering problems [6–10]. With this approach, the search for constants for the equation of state becomes a separate, time-consuming research task. That is why the problem of a few-parameter equation of state has become an important issue.

2. Three-term equation of state

Results of experiments with shock waves provide enough material to construct semi-empirical state equations describing the behavior of condensed matter at high pressures and temperatures. The properties of matter under these conditions are determined by the interaction forces at absolute zero temperature, the thermal vibrations of atoms or ions, and thermal excitation of electrons.

Finding the cold pressure curves and obtaining the equation of state of compressed matter from the results of the dynamic experiments require separation of the pressure and internal



energy into heat and cold components [11]:

$$P = P_c(V) + P_t(V, T), \quad (1)$$

$$E = E_c(V) + E_t(V, T). \quad (2)$$

Elastic components of the pressure and internal energy $P_c(V)$, $E_c(V)$ are associated exclusively with interaction forces acting between the atoms of the body and are equal to the full pressure and specific internal energy at absolute zero, that is why they are sometimes called “cold” pressure or energy. Heat components of pressure $P_t(V, T)$ and energy $E_t(V, T)$ are associated with the body heating, i.e. with the temperature.

The equilibrium state of a solid at absolute zero temperature and zero pressure is characterized by mutual compensation between the atomic forces of attraction and repulsion, as well as the minimum of a potential elastic energy that can be taken as the beginning of its reference $E_c = 0$. At absolute zero temperature, the atoms perform the so-called zero-point oscillations, which are associated with the energy $h\omega/2$ attributed to a single normal oscillation with frequency ω . This energy can be included in potential energy $E_c(V)$.

The elastic pressure is associated with the potential energy through ratio

$$P_c = -\frac{dE_c}{dV}. \quad (3)$$

This ratio (3) makes a natural mechanical sense: the energy gain is equal to the work of compression and can be regarded as equation of isotherm or adiabat of the cold compression. On the other hand, formula (3) follows from the general thermodynamic relation $TdS = dE + PdV$, considering that the temperature T is equal to zero. However, according to the Nernst theorem, entropy S at $T = 0$ is equal to zero as well. Therefore, the isotherm $T = 0$ is isentropic $S = 0$ at the same time.

The cold components do not depend on the temperature and characterize only the elastic interaction of the atoms of the material. The heat components are the reaction of the material to heat. If the temperature is not too high, the atoms of the solid generally oscillate about their equilibrium positions. Moving the atoms in space by hopping in the interstice or other vacant sites requires overcoming potential barriers. During compression, due to the increase of repulsive forces, the potential barrier heights sharply increase as well. Free movements of the particles are yet more complicated, and their movement remains limited by the space of their cells. Under these conditions, it retains the properties of harmonic oscillations in a wide temperature range, including the states arising from the shock compression of “continuous” samples.

The expressions for heat energy $E_{t,l}$ and heat pressure $P_{t,l}$ of the “classical” ensemble of oscillators (lattice atoms) have the form:

$$E_{t,l} = c_{V,l}T, \quad (4)$$

$$P_{t,l} = \gamma_l c_{V,l} \frac{T}{V}, \quad (5)$$

where γ_l is the Grüneisen parameter expressing the ratio of the heat pressure to the density of heat energy, $c_{V,l} = 3R/A$ is specific heat of the lattice at constant volume (the law of Dulong–Petit), A is the average atomic weight, R is gas constant.

Further increase of temperature up to several thousand degrees causes thermal excitation of electrons, primarily, for the metals. Unlike the dielectrics, the metals’ free levels are immediately adjacent to the Fermi surface limiting the filling of the energy states by electrons in the momentum space. Because of this, the equations of state for metals at high temperatures, in addition to the lattice components, should include terms that describe the heat pressure and energy of the electron gas.

When the metal is heated to temperature T , the number of excited electrons is proportional to the number of filled levels νkT (ν is the level density, k is Boltzmann constant) in the energy interval kT . Since the average energy acquired by each electron is proportional to kT , then the total energy of the excited electrons is νT^2 , i.e. in the conventional form [12]

$$E_{t,e} = \frac{c_{V,e}T^2}{2} \quad (6)$$

and, on the basis of the thermodynamic equation $P = T \int_0^T T^{-2}(\partial E/\partial V)dT$, we get

$$P_{t,e} = \frac{\gamma_e c_{V,e}T^2}{2V} \quad (7)$$

where γ_e is an electronic analog of the Grüneisen parameter, expressing the ratio of the heat pressure of electrons to the density of their heat energy, $c_{V,e} = c_{V,e0} (V/V_0)^{\gamma_e}$ is electronic heat capacity, $c_{V,e0}$ is the experimental value of the electronic heat capacity under normal conditions.

To describe the thermal properties of the electron gas, Kormer [12] conducted a detailed analysis of the temperature behavior of electrons based on the quantum statistical calculations by Gilvari [13] and Latter [14]. This analysis showed that below the temperatures of the order of 30000 to 50000 K, the heat capacity of the electrons is proportional to the temperature $c_{V,e} \sim T$, and the energy $E_{t,e} \sim T^2$. As for the thermal pressure, the statistical values of the electronic analog of the Grüneisen parameter γ_e in experimentally attainable range of compressions are constant with great precision and equal to 1/2 (for simple metals), while the high-temperature γ_e for transition metals are about two to three times larger. For the free electron gas $\gamma_e = 2/3$ [11]. In our equations, we use the value $\gamma_e = 2/3$ as well.

Taking into account the expressions (4)–(7), Mie–Grüneisen equations (1) and (2) are reduced to the form

$$E(V, T) = E_c(V) + c_{V,l}T + 1/2c_{V,e0}T^2 \left(\frac{V}{V_0}\right)^{2/3}, \quad (8)$$

$$P(V, T) = -\left(\frac{dE_c}{dV}\right) + \gamma_l \frac{c_{V,l}T}{V} + \frac{1}{3} \frac{c_{V,e0}T^2}{V} \left(\frac{V}{V_0}\right)^{2/3} \quad (9)$$

or, in terms of free energy

$$F(V, T) = E_c(V) + c_{V,l}T \ln\left(\frac{\theta(V)}{T}\right) - 1/2c_{V,e0}T^2 \left(\frac{V}{V_0}\right)^{2/3}, \quad (10)$$

where $\theta(V)$ is the Debye temperature.

3. Construction of the Grüneisen function

Let us construct a model equation of state for thermodynamic functions of solid phase, based on dependence $\gamma(V)$ of the Grüneisen parameter, obtained by Molodets [15,16]

$$\gamma(V) = 2/3 - \frac{2}{(1 - aV_0/V)}, \quad (11)$$

$$a = 1 + \frac{2}{(\gamma_s - 2/3)} + \frac{2P_{T,0}}{K_s}, \quad (12)$$

where $\gamma_s = \beta K_s V_0 / c_V$, β is the coefficient of volume expansion, K_s is adiabatic bulk modulus, c_V is heat capacity at constant volume (in the case of considering the members responsible for

the electronic excitation in equation (10), the heat capacity at constant volume $c_V = c_{V,l} + c_{V,e}$ consists of the lattice and electronic ones), $P_{t,0}$ is the heat part of the pressure under normal conditions.

It should be noted that the procedure for the derivation of equation (11) is “tied” to the initial state (V_0, T_0) , such that $P(V_0, T_0) = 0$. In this thermodynamic state, all thermophysical characteristics $\beta(V_0, T_0)$, $K_S(V_0, T_0)$, $c_V(V_0, T_0)$, $P_{t,0}(V_0, T_0)$ of the material are calculated, which determine parameter a in (12). At that, it should be specifically noted that the derivation is not limited by any assumptions about the type of condensed matter, and the formula itself contains only general fundamental properties of the material.

Using the definition of the Grüneisen parameter in the Debye approximation

$$\gamma = - \left(\frac{d \ln \theta}{d \ln V} \right)$$

and (11), we find the dependence of the Debye characteristic temperature on the volume

$$\theta(V) = \theta_0 \left[\frac{(a - V/V_0)}{(a - 1)} \right]^2 \left(\frac{V_0}{V} \right)^{2/3}, \quad (13)$$

where $\theta_0 = \theta(V_0)$ is the value of Debye temperature under normal conditions.

4. Finding the zero isotherm

Let us find the dependence of the Grüneisen parameter on the volume $\gamma(V)$. According to Slater [17] and Landau [18], all the frequencies vary in proportion to the speed of sound $C = V(-dP_c/dV)^{1/2}$ and are inversely proportional to the interatomic distance $r \sim V^{-1/3}$. Under these assumptions, the average rate for the “classical” ensemble of oscillators

$$\omega_S \sim V^{2/3} \left(-\frac{dP_c}{dV} \right)^{1/2}. \quad (14)$$

The average rate by Dugdale and McDonald model, according to [19], is calculated as follows:

$$\omega_{DM} \sim V^{1/3} \left[-\frac{d}{dV} (P_c V^{2/3}) \right]^{1/2}. \quad (15)$$

Model of Zubarev and Vaschenko [20] for the particles, vibrating in a spherically symmetric field of its neighbors, gives average rate

$$\omega_{ZV} \sim \left[-\frac{d}{dV} (P_c V^{2/3}) \right]^{1/2} \quad (16)$$

Comparison of the calculated dependences $\gamma(V)$ obtained in different ways (see equations (14)–(16)) with the experimental data shows that none of the quasi-harmonic models adequately describes the dynamic compression [2]. Therefore, for the most common expressions for the potential energy on the zero isotherm, all the different assumptions can be combined into a single formula for the mean frequency

$$\omega \sim V^{\frac{2-t}{3}} \left[-\frac{d}{dV} (P_c V^{\frac{2t}{3}}) \right]^{1/2}. \quad (17)$$

It is its logarithmic derivative by the volume that determines the generalized Grüneisen parameter

$$\gamma(V) = -\left(\frac{2-t}{3}\right) - \frac{V}{2} \left[\frac{\frac{d^2}{dV^2} \left(P_c V^{\frac{2t}{3}} \right)}{\frac{d}{dV} \left(P_c V^{\frac{2t}{3}} \right)} \right]. \quad (18)$$

Then, the value in (18) at $t = 0$ corresponds to the theory of Landau–Slater, $t = 1$ —Dugdale–Macdonald, $t = 2$ corresponds to the free volume theory. It should be noted that the magnitude and sign of parameter t is determined by the nature of Poisson’s ratio. Zero value of parameter t corresponds to constant Poisson’s ratio, i.e. to Slater’s formula (14). A positive value of t corresponds to an increase of Poisson’s ratio with the pressure, i.e. to formulas of Dugdale–MacDonald (15) and Zubarev–Vaschenko (16). Finally, a negative value of t corresponds to a negative derivative of Poisson’s ratio.

In the case of normal conditions at $V = V_0$, we have the following relation for the value of the Grüneisen parameter:

$$\gamma|_{t=0} = \gamma|_{t=1} + \frac{1}{3} = \gamma|_{t=2} + 2/3.$$

Comparison of different methods of calculating $\gamma(V)$ with the experimental data for aluminum and copper in [21] has shown that none of the quasi-harmonic models (14)–(16) provides a decisive advantage in the description of the dynamic experiment, therefore, in general, the use of equation (17) is the most reliable. Additionally, we note that the use of formula (18) for the calculation of $\gamma(V)$ is acceptable, strictly speaking, only for isotropic structures or those having cubic symmetry, while in the general case, it is necessary to take into account the tensor nature of the Grüneisen parameter.

To find the zero isotherm, we equate correlations for the Grüneisen parameter at zero temperature $T = 0$ from equations (11), (12) and the expression for the generalized Grüneisen parameter (18). Then, we can obtain the differential equation for the “cold” pressure P_c

$$2/3 - \frac{2}{1 - a_c V_0/V} = -\left(\frac{2-t}{3}\right) - \frac{V}{2} \left[\frac{\frac{d^2}{dV^2} \left(P_c V^{\frac{2t}{3}} \right)}{\frac{d}{dV} \left(P_c V^{\frac{2t}{3}} \right)} \right], \quad (19)$$

where a_c is the value of parameter $a|_{T=0}$ at zero temperature in equation (12). According to equation (11), $a_c = a(0) = 1 + 2/(\gamma_s - 2/3)$ can be taken as the first approximation.

The solution of differential equation (19) allows one to determine the “cold” pressure and energy

$$P_c(V) = C_1 V^{-2t/3} + C_2 H_2(V), E_c(V) = -\left(\frac{C_1 V^{(1-\frac{2t}{3})}}{1 - \frac{2t}{3}} + C_2 H_1(V) \right) + C_3 \quad (20)$$

where C_1 , C_2 and C_3 are integration constants, and $H_1(V)$ and $H_2(V)$ are polynomials computed by the following formulas [22, 23]

$$H_1(V) = V^{-2/3} \left(-\frac{3}{2} k_1 \xi^4 - 12 k_2 \xi^3 V + \frac{9}{2} k_3 \xi^2 V^2 - \frac{12}{7} k_4 \xi V^3 + \frac{3}{10} k_5 V^4 \right);$$

$$H_2(V) = V^{-\frac{5}{3}} (k_1 \xi^4 - 4 k_2 \xi^3 V + 6 k_3 \xi^2 V^2 - 4 k_4 \xi V^3 + k_5 V^4).$$

The following replacements are made here:

$$\xi = a_c V_0, \quad k_1 = t_2 t_{12} t_1 t_{72}, \quad k_2 = t_2 t_{12} t_{52} t_{72}, \quad k_3 = t_2 t_1 t_{52} t_{72}, \quad (21)$$

$$k_4 = t_{12} t_1 t_{52} t_{72}, \quad k_5 = t_2 t_{12} t_1 t_{52}, \quad (22)$$

$$t_2 = t + 2, \quad t_{12} = t + 1/2, \quad t_1 = t - 1, \quad t_{72} = t + 7/2, \quad t_{52} = t - 5/2. \quad (23)$$

To find the constants in (20), we perform the following steps, similar to [15].

- (i) Having substituted equation (20) into the equation for the pressure (8) under normal conditions $T = T_0$ $V = V_0$, we obtain point at isotherm of the normal state

$$P(V_0, T_0) = C_1 V_0^{-2t/3} + C_2 H_2(V_0) + \frac{\gamma c_{V,t} T_0}{V_0} + \frac{c_{V,e0} T_0^2}{3V_0}. \quad (24)$$

Having differentiated isotherm (20) by volume V and using the experimental value of the bulk isothermal compression modulus K_t under normal conditions

$$C_1 = 3V^{(1+\frac{2t}{3})} \frac{\left[\left(\frac{\partial H_2(V)}{\partial V} \right) (P_0 - P_{t,0}) + H_2(V) \left(\frac{K_t}{V_0} + \left(\frac{\partial P_{t,0}}{\partial V} \right) \Big|_T \right) \right]}{2t H_2(V) + 3V \left(\frac{\partial H_2(V)}{\partial V} \right)}. \quad (25)$$

- (ii) Since $P(V_0, T_0) = P_0$, then, having substituted expression (25) into (24), we obtain the value of constant C_2

$$C_2 = \frac{\left[2t (P_0 - P_{t,0}) + 3V \left(\frac{K_t}{V_0} + \left(\frac{\partial P_{t,0}}{\partial V} \right) \Big|_T \right) \right]}{2t H_2(V) + 3V \left(\frac{\partial H_2(V)}{\partial V} \right)}, \quad (26)$$

where $P_{t,0}$ is thermal pressure under normal conditions, corresponding to the second and third terms in the equation for the pressure (8).

- (iii) In order to find constant C_3 , we begin with finding the root of equation $P_c(V_*) = 0$, which is obtained from equation for the pressure (20). Then, we assume that at the specific volume equal to $V_{0,V} = V_0 a_c V_*$, the potential energy is zero (potential well)

$$C_3 = \frac{C_1 V_*^{(1-2t/3)}}{1 - 2t/3} + C_2 H_1(V_*). \quad (27)$$

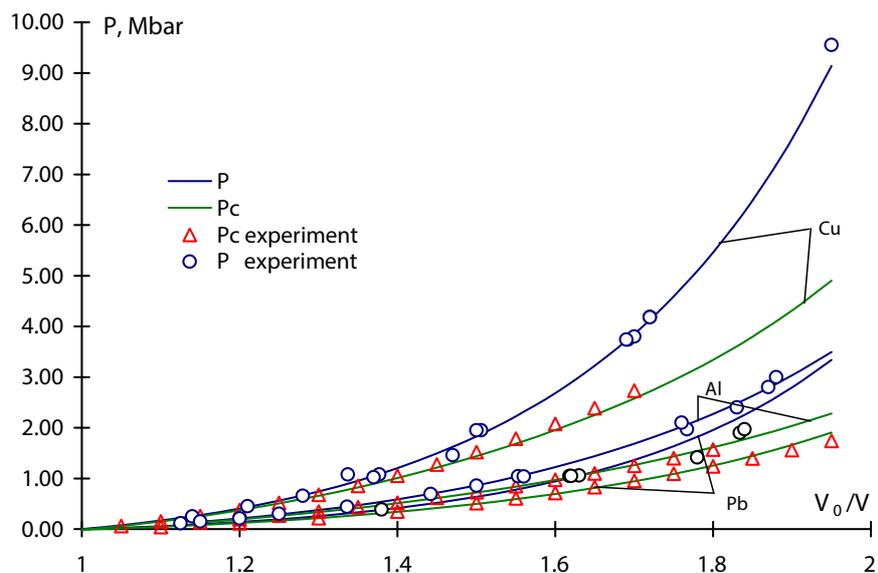
5. The scope of applicability of the thermodynamic model

One of the important moments in the development of the thermodynamic equations of state is a question of the scope of their applicability. Parameters V_0 , β , K_t , c_P , Θ_0 , $c_{V,e0}$ of the model are determined from the reference data for particular initial conditions, i.e., they characterize only that phase of the matter, in which the substance exists under given initial conditions (one phase of the substance) (see table 1).

Another limitation of the model is that relation (11) has a singularity at value of the current volume $V = a_c V_0$. According to expression, it corresponds to the zero value of the characteristic temperature, so at this point, all the thermodynamic functions of the material are discontinued. Since this point lies in the area of strains, the correct application of the model in the area of strains requires the use of other equations of state with conditions of crosslinking under normal conditions, which is the subject of a separate study.

Table 1. The thermodynamic parameters of materials.

| | Θ_0 , K | β , 10^{-6} K | V_0 , cm^3/g | K_t , GPa | c_P , J/mol K | $c_{V,e0}$, mJ/mol K^2 |
|-------------------------|----------------|-----------------------|--------------------------------|-------------|-----------------|----------------------------------|
| Al [24] | 433 | 69.9 | 0.372 | 72.2 | 24.35 | 1.35 |
| Cu [24] | 347 | 50.1 | 0.112 | 137.1 | 24.43 | 0.69 |
| Pb [24] | 105 | 85.5 | 0.088 | 43.2 | 26.65 | 2.99 |
| UO ₂ [25,26] | 385 | 29.2 | 0.097 | 208 | 62.6 | — |

**Figure 1.** Principal Hugoniot and curves of “cold” compression of aluminum (Al), copper (Cu), and lead (Pb).

6. The calculation results

To calculate the behavior of condensed matter behind the front of shock waves, the original expressions need to be supplemented with the Rankine–Hugoniot relation [11]

$$E_H = E_0 + 1/2P_H(V_0 - V),$$

that allows of calculating all the thermodynamic parameters of materials on principal Hugoniot. Table 2 summarizes the parameters of the model sufficient to calculate the cold energy and pressure according to the formula (15). Figure 1 shows the principal Hugoniot and zero isotherms of aluminum, copper, and lead calculated by the authors’ method.

For comparison, the results of the experiments of various research groups are shown, which data are put together in [27], as well as the results of calculation of the “cold” pressure [28] (these calculations are classical in the area of creation of the equation of state). As one can see, the differences in the “cold” curves calculated in [28] and the authors’ dependences are insignificant up to the compression ratios $V_0/V = 1.5$. At the compression ratios $V_0/V > 1.5$ the differences are more significant (but they do not exceed 7%) due to the method of constructing the model, where the reference state for the construction of equations is the normal state of condensed matter.

Table 2. The parameters of the model.

| | t | a_c | C_1 , GPa | C_2 , GPa | C_3 , kJ/g |
|-----------------|-----|-------|-------------|-------------|--------------|
| Al | 0.0 | 2.62 | 22.2 | -0.19 | -1091 |
| Cu | 0.0 | 2.34 | 69.7 | -13.02 | -9245 |
| Pb | 0.0 | 2.01 | 42.7 | -19.8 | -3810 |
| UO ₂ | 0.6 | 4.98 | 4.7 | -0.3 | -1132 |

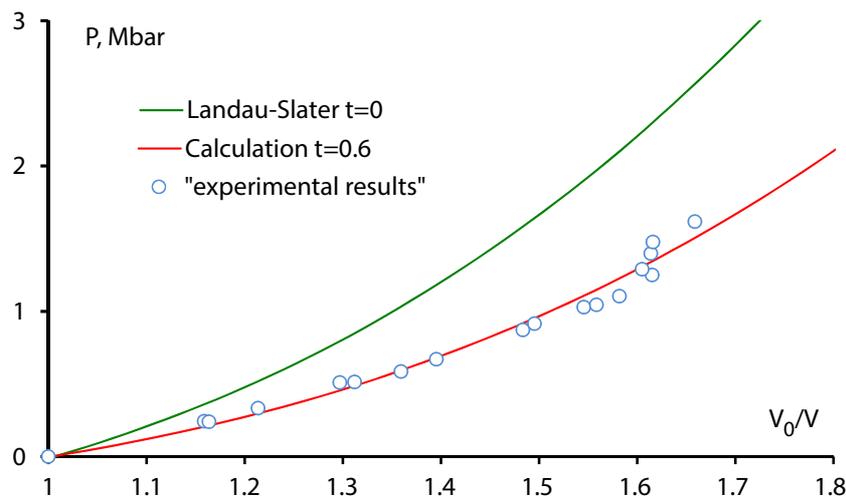


Figure 2. Principal Hugoniot of UO₂.

The calculations were conducted at parameter value $t = 0$ in equation (18), i.e. by the theory of Landau–Slater. Calculations for the uranium dioxide UO₂ have shown that there are no values of the fitting parameter a_c or the initial data, which determine the initial course of the principal Hugoniot, that would allow of approximating the experimental data from [29] at $t = 0$. For $t > 1$ the approximation is possible, but derivative $\partial P/\partial \rho$ at the starting point for these experiments is quite small. Figure 2 shows the calculation of the shock Hugoniots calculated at $t = 0$, by the Landau–Slater method and at $t = 0.6$ (18). The use of the Grüneisen parameter in the generalized form allowed of achieving a good match of the numerical simulation [8] and the experimental data.

7. Conclusion

The number of semi-empirical relations (10), (13), (20) allows one to describe the behavior of the thermodynamic properties of solids in a wide range of pressures and temperatures. At that, to use the equation of state, it is necessary and sufficient to know only eight constants. V_0 , β , K_t , c_P , Θ_0 , $c_{V,e0}$ corresponding to the values of the variables under normal conditions, which can be found in directories on the physical and mechanical properties of materials [27]. Parameter a_c , which is also included in the equation of state, is to be specified from the experimental data (e.g., isotherm or Hugoniot) for a given value of parameter t . The remaining values c_V and K_s , which are also used in the equations for the calculations, are determined from the above

parameters using known thermodynamic identities

$$K_s = K_t + \frac{TV(K_t\beta)^2}{c_V}, \quad c_V = c_P - TVK_t\beta^2.$$

Thus, a simple caloric model of the equation of state is proposed to solve the high-speed dynamic problems [8,30,31], providing accuracy comparable to that of the wide-range equations of state in the area of compressions under consideration, with the minimal number of constants needed for the solution.

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