Possibility of interpolating the Debye approximation to describe liquid and gas phases

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(Submitted 17 May 1995)
Zh. Éksp. Teor. Fiz. **100**, 162–173 (January 1996)

A method for interpolating the Debye approximation, which describes the crystalline state of a substance, to the liquid and gaseous regions is described. The method formally reduces to multiplying the Debye temperature by the factor $1+L$, where L is proportional to the ratio of the mean amplitude of thermal vibrations of the atoms in the substance to the mean interparticle distance (the Lindemann parameter). In the crystalline state $L \ll 1$, but $L \to \infty$ upon transition to the gas. Thermodynamics with a Debye temperature renormalized in such a manner, i.e., a generalized Debye approximation, which describes the solid state and the ideal gas in limiting cases, is constructed. It is shown that under this approximation the dependence of the Debye temperature on the volume and the corresponding Grüneisen coefficient can be found from the condition that the calculated values of the entropy on the coexistence curves of different phases correspond to the tabulated values. It is thereby possible to obtain a unified description of the different states of a substance. The thermodynamics of neon is considered. The special features of the behavior of the thermal and elastic components of the pressure upon transition from the liquid to the solid are discussed. It is shown that the constancy of the known Lindemann parameter in the melting region is attributable to equality between the thermal components of the pressure of the solid and liquid phases during melting. Surfaces of state are constructed for the solid and fluid phases. © *1996 American Institute of Physics.* [S1063-7761(96)00901-2]

1. INTRODUCTION

Thermodynamic functions, which realistically describe the characteristics of a substance in different parts of its phase diagram, are fundamental characteristics of matter. The use of such functions has always been appropriate and is constantly growing. At the present time, empirical methods have achieved complete dominance in this area. For example, the modern broad-range equations of state, which were derived to describe metals over a broad range of compression parameters, contain tens of free and empirical parameters.[1–5] The latter are determined on the basis of data from shock-wave experiments, measurements of the release isentropes of porous samples, and other experimental thermodynamic information over a broad region of the phase diagram.

At the same time, since the monumental work of van der Waals, the attempts to construct the thermodynamics of a substance by interpolating known thermodynamic functions from different regions of the phase diagram have never ceased. The Debye interpolation formula, which describes the state of a solid in the high- and low-temperature limits, is well known.[6,7] In the melting region the coexistence curves are determined on the basis of the Lindemann method,[7] from which an equation similar to Simon's empirical equation is derived. However, these methods are valid only over restricted portions of the phase diagram.

In this paper we propose an interpolation that extends the Debye approximation into the region of the liquid and the gas. This is achieved by introducing an effective Debye temperature, which differs from the ordinary Debye temperature by multiplication by a certain function that depends on a parameter similar to the familiar Lindemann parameter. In the solid-state region the correction associated with this function is small. However, when we move into the gaseous region, it increases and provides for conversion of the thermodynamic functions of the solid into those of the ideal gas. The thermodynamic functions formally have the form of Debye functions, which have been supplemented by terms associated with the temperature dependence of the effective Debye temperature. The thermodynamics thus constructed, which we call a generalized Debye approximation, contains one determinable dependence, viz., the dependence of the Debye temperature on the volume. The latter can be determined from experimental or theoretical data, and once it is found, the thermodynamics of the substance will have been constructed over a broad range of states.

The thermodynamics of the inert gas neon is considered as an example. The tabulated theoretical data in Ref. 8, which were obtained with consideration of the entire set of experimental results available at that time, are used. The values of the entropy on the coexistence curves taken from Ref. 8 are set equal to the expression for the entropy following from the thermodynamics considered here. The equation thus obtained is used to determine the Debye temperature as a function of the volume in the region of states corresponding to the cold solid, through the melting region, and in the region corresponding to the ideal gas. The Grüneisen coefficient is found from it by logarithmic differentiation.

As a result of the calculation, it is found that the transition from the solid to the liquid state is accompanied not only by a discontinuous change in the specific volume, but also by an abrupt decrease in the Debye temperature. This circumstance was noted back in Ref. 9. The Debye temperature

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decreases monotonically from the liquid to the gaseous state. When the dependence of the Debye temperature on the volume is known, any thermodynamic function can be calculated using the generalized Debye approximation. The behavior of the Lindemann parameter on the coexistence curves is interesting. In particular, it remains practically constant on the solidification and melting curves, in agreement with Lindemann's original hypothesis. In the generalized Debye approximation this is a consequence of the constancy of the pressure upon melting.

The elastic (cold) and thermal components of the pressure on all the coexistence curves are calculated. The variation of these components upon passage from the crystalline to the liquid state and from the liquid to the gaseous state is discussed. Surfaces of states are constructed in the volume-temperature plane, and the phase equilibrium curves are shown.

2. EFFECTIVE DEBYE TEMPERATURE

Let us consider the thermal component of the free energy of a solid in the Debye approximation for high temperatures:

$$F=\frac{RT}{\mu}\ln\left(\frac{\Theta_D}{T}\right)^3. \tag{1}$$

Here R is the universal gas constant, μ is the atomic weight, T is the temperature, and

$$\Theta_D=\Theta_0\exp\left(-\int_{v_0}^{v}\Gamma_s d\ln v\right), \tag{2}$$

where Θ_0 is the Debye temperature at $T=0$, at which the specific volume of the gas $v=v_0$, and Γ_s is the Grüneisen coefficient. We note that the expression (1) holds when $\Theta_D/T\ll 1$.

We write the thermal component of the free energy of an ideal gas:

$$F_g=\frac{RT}{\mu}\left(\ln\frac{\Lambda^3}{g_a mv}-1\right), \tag{3}$$

where g_a is the partition function of a gas atom and $\Lambda^3=h^3/(2\pi mkT)^{3/2}$ is the thermal de Broglie wavelength for a particle of mass m. We note that Eq. (3) can be rewritten in the form

$$F_g=\frac{RT}{\mu}\left[\ln\left(\frac{T_F}{T}\right)^{3/2}-1\right], \tag{4}$$

where

$$T_F=\frac{h^2}{(2\pi mk)(g_a mv)^{2/3}} \tag{5}$$

is actually the Fermi temperature. It differs from the exact ratio determined from the condition $kT_F=E_F$ (E_F is the Fermi energy) by an insignificant numerical factor.

We introduce the effective Debye temperature

$$\Theta=\Theta_D+\sqrt{T_F T}, \tag{6}$$

and note that the following inequality holds at temperatures and densities corresponding to the solid state:

$$\Theta_D\gg\sqrt{T_F F}. \tag{7}$$

This means that in the solid state $\Theta\cong\Theta_D$. However, as the temperature T and the volume v increase, the second term in (6) begins to dominate (Θ_D decreases exponentially with increasing volume, and $\sqrt{T_F T}$ decreases only as $1/v^{1/3}$).

We also note that (6) can be represented in the form

$$\Theta=\Theta_D(1+L), \tag{8}$$

where

$$L=\frac{\sqrt{T_F T}}{\Theta_D}=\frac{h\sqrt{T}}{\sqrt{2\pi mk}(g_a mv)^{1/3}\Theta_D}. \tag{9}$$

The quantity mv equals n, i.e., the concentration of the substance, and $n^{1/3}$ is of the order of the mean distance r between atoms. Therefore, L^2, which is the ratio between the mean-square amplitude of thermal vibrations and the square of the mean interparticle distance, is a natural generalization of the familiar Lindemann parameter from the theory of melting.[7] Lindemann considered the ratio between the mean-square amplitude of the thermal vibrations and the square of the lattice constant. In the solid state $L\ll 1$. The value of L increases upon transition to the gas.

As we know, the logarithmic derivative of Θ with respect to the volume defines the Grüneisen coefficient. Using (6), we obtain an expression for the effective Grüneisen coefficient

$$\Gamma=\frac{\Gamma_s+L/3}{1+L}. \tag{10}$$

It is seen that when $L\to\infty$, the value of Γ tends to the "ideal-gas" value of 1/3.

We substitute the effective Debye temperature thus introduced into the expression for the thermal component of the free energy in the Debye approximation:

$$F=\frac{RT}{\mu}\left\{3\ln\left[\frac{\Theta_D}{T}(1+L)\right]-D\left(\frac{\Theta_D}{T}(1+L)\right\}, \tag{11}$$

where

$$D(x)=\frac{3}{x^3}\int_0^x\frac{y^3dy}{e^y-1} \tag{12}$$

is the Debye function. As $L\to\infty$, $D\to 1$, and Eq. (11) transforms in Eq. (3) for an ideal gas. All the quantities in it, except the function $\Theta_D(v)$, are known. We determine the latter from the condition that the thermodynamic functions obtained on the basis of (11), correspond to the most accurate presently available tabulated data. The latter are consistent with the entire body of existing experimental data. This is the main idea of the interpolation described in this paper.

3. GENERALIZED DEBYE APPROXIMATION

We formally introduce the effective Debye temperature and an expression for the total free energy in the Debye approximation:

$$F=\frac{RT}{\mu}\left[3\ln(1-e^{-\Theta/T})-D\left(\frac{\Theta}{T}\right)\right]-U(v), \tag{13}$$

where U is the elastic (cold) component of the energy at $T=0$. We call the free energy in the form (13) and the thermodynamic functions following from it a generalized Debye approximation. We obtain the latter from (13) on the basis of the ordinary equations of thermodynamics, taking into account the dependence of the effective Debye temperature on the volume and the temperature. Then the expression for the entropy has the form

$$S=\frac{R}{\mu}\left\{4D\left(\frac{\Theta}{T}\right)\left[1-\frac{3L}{8(1+L)}\right]-3\ln(1-e^{-\Theta/T})\right\}. \quad (14)$$

When L is large, it is easy to show that (14) corresponds exactly to the expression for an ideal gas:

$$S=\frac{R}{\mu}\left[\frac{5}{2}-\ln\frac{h^3}{(2\pi mkT)^{3/2}g_a mv}\right]. \quad (15)$$

The energy is

$$E=\frac{3RT}{\mu}D\left(\frac{\Theta}{T}\right)\left[1-\frac{L}{2(1+L)}\right]. \quad (16)$$

It equals $3RT/\mu$ at small L and $3RT/2\mu$ at large L, i.e., a transition to an ideal gas occurs.

The pressure is

$$p=\frac{3RT}{\mu v}D\left(\frac{\Theta}{T}\right)\frac{\Gamma_s+L/3}{1+L}-\Pi(v), \quad (17)$$

where

$$\Pi(v)=-\frac{dU(v)}{dv} \quad (18)$$

is the elastic (cold) component of the pressure. As we see, the pressure is written in the Mie–Grüneisen form. At large L and T Eq. (17) gives the ideal gas pressure.

The specific heat at constant volume is

$$C_v=\frac{3R}{\mu}\left[4D\left(\frac{\Theta}{T}\right)-\frac{3\Theta/T}{e^{\Theta/T}-1}\right]\left[1-\frac{L}{4(L+1)}\times\left(3-\frac{L}{L+1}\right)\right]. \quad (19)$$

At low temperatures, at which $\Theta/T\gg 1$, we have $C_v\propto T^3$. At high temperatures, at which $\Theta/T\ll 1$ and $L\ll 1$, we have $C_v=3R/\mu$. Finally, at high temperatures and large values of L, the specific heat becomes equal to the specific heat of an ideal gas $C_v=3R/2\mu$.

Thus, we can pass from the thermodynamics of a solid to the thermodynamics of an ideal gas using the effective Debye temperature without introducing any undetermined interpolation parameters. Equations (13)–(19) contain the function $\Theta_D(v)$ and the corresponding Grüneisen coefficient Γ_s, as well as the elastic component of the pressure. Once these functions are found, the thermodynamics is thereby constructed over a fairly extensive region of the phase diagram. We note that these functions must be determined on the basis of a variety of considerations when the ordinary Debye approximation is used. In this sense the generalized Debye approximation does not raise new problems. The only difference is that in the latter case $\Theta_D(v)$ and $\Pi(v)$ must be determined over a broader range of variation of v.

4. DETERMINATION OF THE DEBYE FUNCTION AND THE GRÜNEISEN COEFFICIENT

We determine the unknown function $\Theta_D(v)$ from the condition that the thermodynamics constructed on the basis of Eqs. (13)–(19) corresponds to the tabulated data for the thermodynamic functions of various substances. We choose the inert gas neon as an object. Its thermodynamic properties have been thoroughly studied and were tabulated in Ref. 8. The tables in Ref. 8 were obtained by theoretical means and were reconciled with the entire body of experimental data on the thermodynamic properties of inert gases.

Since the thermodynamics developed above has the correct asymptotes, its correspondence to the tabulated data on the coexistence curves is most important. The tables in Ref. 8 give the entropy on the melting, solidification, vaporization, and condensation curves. Unfortunately, there are no values of the entropy on the sublimation curve. Only values of the entropy in the crystalline state as a function of the temperature at constant pressure are given. However, in that state the entropy is a very weak function of the pressure. For example, when the pressure is varied from 1 to 100 atm, the difference in the entropy values of xenon taken at $T=T_c$ amounts to 0.5%. This suggests that the values of the entropy on the sublimation curve are virtually independent of the pressure. We used the data for $p=1$ atm.

The tabulated entropy value for specific values of T and v along a coexistence curve was set equal to the expression following from Eq. (14). The value of $\Theta_D(v)$ was found numerically from the equation thus obtained. This procedure was carried out for all five coexistence curves. The results are presented in Fig. 1, which shows the dependence of Θ_D on v/v_c (v_c is the specific volume at the critical point). Figure 1(a) contains the coexistence curves for the solid phase, viz. the sublimation curve (curve *1*) and the solidification curve (curve *2*), as well as the melting curve (curve *3*) for the liquid phase. As is seen from Fig. 1(a), the values of the Debye temperatures on the sublimation and solidification curves differ somewhat. Such a disparity is not surprising, since the sublimation curve was drawn according to the $p=1$ atm isobar and the accuracy of the tabulated data is not high enough to detect volume changes at very low temperatures. Apparently, the high-temperature portion of the solidification curve is also known to poor accuracy. Curve *3* corresponds to the liquid phase. The transition from the solidification curve to the melting curve is accompanied by abrupt changes in the volume and the Debye temperature. This corresponds to the idea advanced in Ref. 9 that the transition from the solid to the liquid should be accompanied not only by an abrupt increase in volume, but also by a certain decrease in the mean vibrational frequency, i.e., the Debye temperature. The discontinuity with respect to v in Fig. 1(a) corresponds to the portion of the melting curve at high temperatures and pressures, for which there are no data in the tables in Ref. 8.

Plots of $\Theta_D(v/v_c)$ for the liquid and gaseous branches of the coexistence curves are shown in Fig. 1(b). Vaporiza-

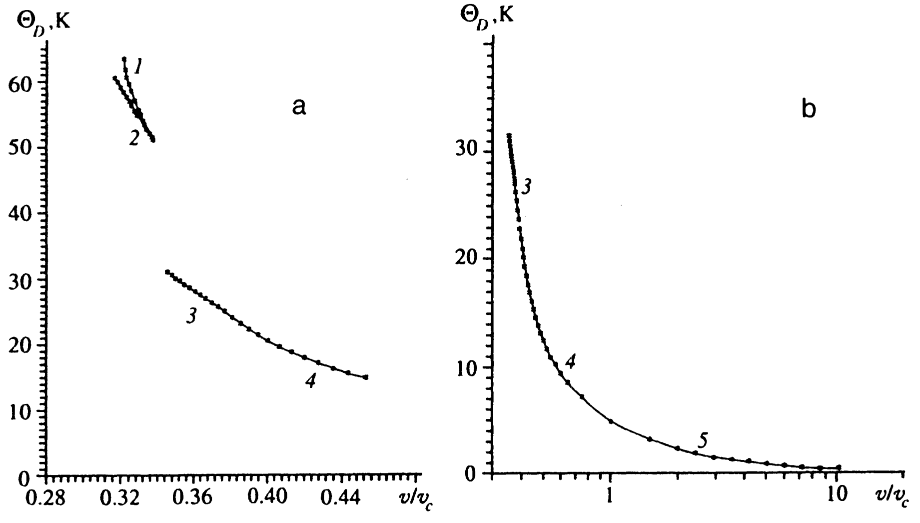

FIG. 1. Dependence of the Debye temperature Θ_D on v/v_c: *1*) sublimation curve; *2*) solidification curve; *3*) melting curve; *4*) vaporization curve; *5*) condensation curve. The same numbering is used in the subsequent figures.

tion curve *4* and condensation curve *5* have been added to melting curve *3*. As is seen, the curves pass smoothly into one another, and Θ_D decreases monotonically with increasing v.

These calculations demonstrate the feasibility of using a single dependence of Θ_D to describe different portions of the phase diagram. The slight ambiguity in the determination of Θ_D in the solid state is probably due to insufficient accuracy of the existing data on the solid phase at low values of v.

In Fig. 2 Θ_D and Θ (dashed curve) are plotted as functions of the temperature for all the coexistence curves. As is seen, the Debye temperature increases with increasing volume along the solidification curve. The transition to the fluid branches is discontinuous. Experimental low-temperature values of the Debye temperature,[10] which were obtained by processing the isochoric values of the specific heat for the volume corresponding to $T=0$, are also plotted in this figure. As is seen from the figure, the experimental values lie near our plot of Θ_D. It should also be noted that the differences between Θ_D and Θ are within the range of error of the experimental data.

The plots of $\Theta_D(v)$ found were used to construct plots of $\ln[\Theta_D(v)]$ as a function of $\ln v$, and the values of the Grüneisen coefficient were found by numerical differentiation. The results are presented in Figs. 3 and 4. On the sublimation and solidification curves we have $\Gamma_s \cong 2.67$. In the liquid state the Grüneisen coefficient becomes slightly smaller on the melting curve ($\Gamma_s \cong 2.57$), and then it decreases smoothly on the vaporization curve, continuing along the condensation curve at $\Gamma_s \cong 1.17$.

The effective Grüneisen coefficient defined by (10) varies more smoothly. It is plotted as dashed lines for the corresponding phase regions in Fig. 3.

5. INVESTIGATION OF THE THERMODYNAMICS. DETERMINATION OF THE ELASTIC COMPONENT OF THE PRESSURE

The Lindemann parameter L on the equilibrium curves is plotted as a function of T in Fig. 4. On the sublimation curve

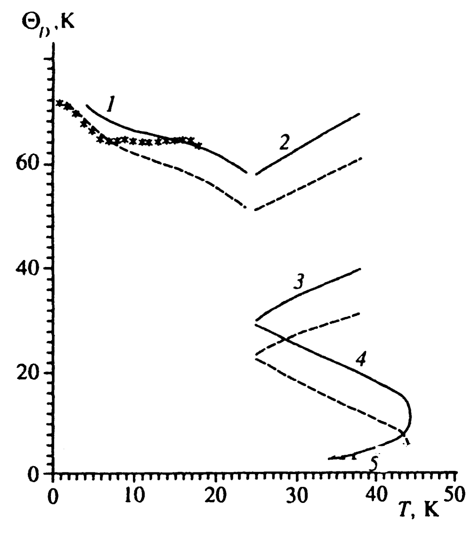

FIG. 2. Debye temperature for Ne: experiment in Ref. 10 (asterisks); calculations in the present work for Θ_D (solid curves) and Θ (dashed curves).

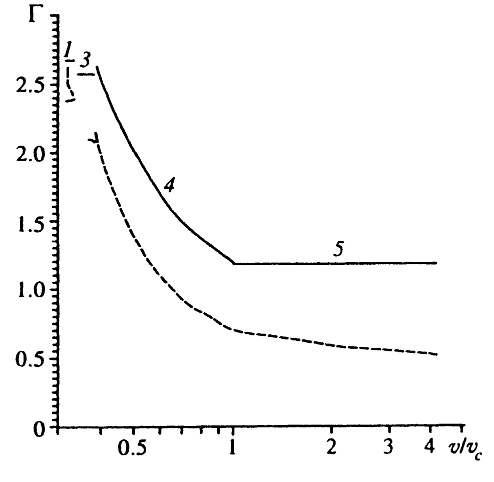

FIG. 3. Grüneisen coefficient. The dashed curve was calculated from Eq. (10).

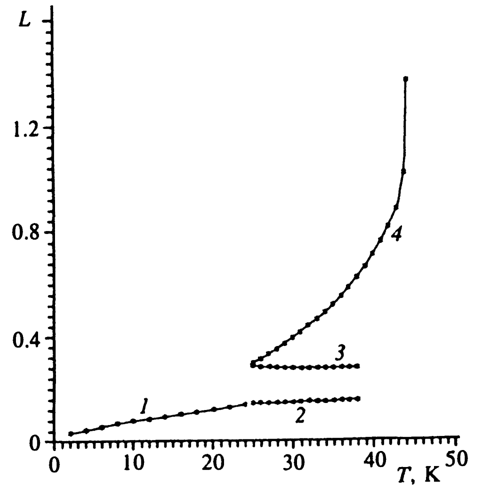

FIG. 4. Temperature dependence of the Lindemann parameter on the coexistence curves.

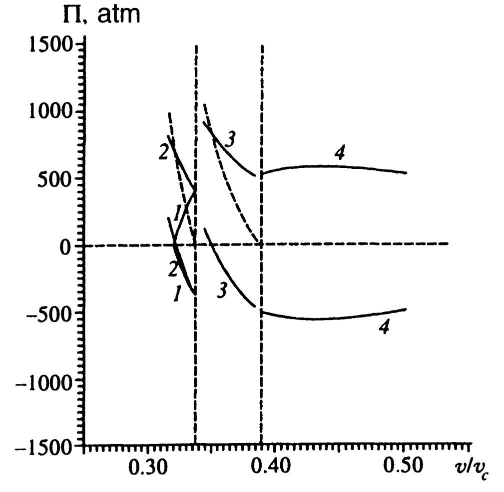

FIG. 6. Thermal and elastic components of the pressure on the coexistence curves. Dashed lines—pressure in the melting region.

it increases from small values to 0.1 at the temperature of the triple point. The value of L remains essentially constant on the solidification and melting curves, in agreement with Lindemann's hypothesis.[7] The value of L increases again on the vaporization curve, reaching values of the order of unity at the critical point. On the gaseous branch of the binodal (*5*) (Fig. 5) L increases rapidly, and a transition to ideal gas thermodynamics takes place. The reason for the constancy of L in the melting region will be discussed below.

After determining Θ_D and Γ_s, we can easily calculate any thermodynamic quantity from (13)–(19). Figure 5 presents the values of the specific heat C_v on the phase equilibrium curves. The numbering of these curves is the same as in the preceding figures. It is seen that the specific heat C_v increases on the sublimation curve according to a $C_v \propto T^e$ law. The behavior of C_v on the solidification and melting curves is peculiar. The value of C_v decreases smoothly along the vaporization curve from a peak value of $\sim 2R/\mu$, passing onto the condensation curve after the critical point and tending to a value of $3R/2\mu$, which corresponds to an ideal gas.

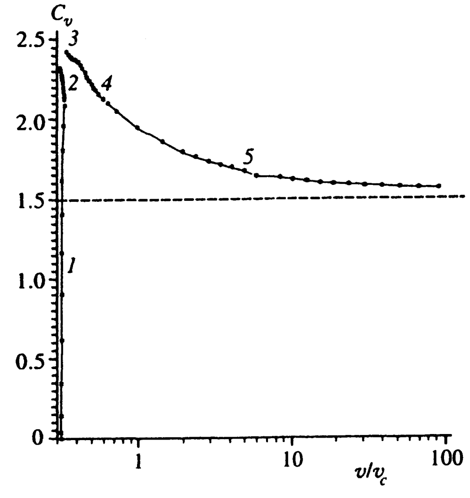

FIG. 5. The specific heat C_v (in units of R/μ) on the coexistence curves.

It should be noted that C_v never takes the value $3R/\mu$, which corresponds to the high-temperature limit of the solid. This is due to the fact that the Debye temperature Θ_0 for Ne is 74 K, while the critical temperature is $T_c=44.4$ K. The temperature at the triple point for inert gases is $T_t=0.556T_c$. All this results in the fact that the high-temperature limit of the Debye theory $\Theta_D/T \ll 1$ is not realized in Ne.

It would be interesting to use the tabulated data in Ref. 8 and the values found for Θ_D and Γ_s to obtain information on the relation between the thermal and elastic components of the pressure in different portions of the phase diagram. The elastic component of the pressure can be calculated from (17) if the tabulated values are used for p. We note that the resultant pressure on the sublimation and vaporization curves is small compared with the thermal and elastic components. Thus, we have

$$\Pi(v) \cong \frac{3RT}{\mu v} D\left(\frac{\Theta}{T}\right) \frac{\Gamma_s + L/3}{1+L}. \tag{20}$$

In the melting region the exact equation (17) should be used to perform the calculation.

The thermal and elastic components of the pressure (in atm) as functions of the specific volume in the form of v/v_c are presented in Fig. 6 for different states. The vertical dashed lines in the figure correspond to the volumes of the solid and liquid phases at the triple point $v_{ts}/v_c=0.336$ and $v_{tl}/v_c=0.387$. On sublimation curve *1* the thermal and elastic components are close to one another. The difference between them, which is equal to the pressure on the sublimation curve, is small compared with the values of the thermal and elastic pressures. Curves *2* were constructed from the data on the solidification curve. As expected, the cold component determined from these data nearly coincides with that found from the sublimation curve. The thermal component decreases with increasing volume. At high pressures and small volumes it greatly exceeds the elastic pressure, so that the pressure on the solidification curve becomes essentially equal to the thermal pressure. We note that the highest possible specific volume for the solid state is achieved at the triple point. The solidification of inert gases is associated with a decrease in the specific volume. This property is not universal, but is typical of the type of substance under consideration, in which the solidification and melting curves are "tilted" toward smaller volumes. As we know, the situation in the case of water is different.

Curves *3* were constructed from the data on the melting curve. As we see, they are similar to curves *2* to some extent, being only displaced toward larger volumes by an amount corresponding to the jump in the volume at the triple point. This graphically supports the familiar idea of abrupt passage from the crystal to the liquid. Curves *4* were constructed on the basis of the vaporization curves. Curves *5*, which are smooth extensions of curves *4*, are not shown in Fig. 6. "Cold" curves *3* and *4* reach a minimum and then descend smoothly, tending to zero in the range of negative pressure values.

We note that in curves *4* and their extensions *5* up to the critical point, the difference between the thermal and elastic pressure which determines the resultant pressure on the vaporization curve, is also small compared with these pressures.

Some important conclusions regarding the behavior of the substance in the melting region can be drawn from the curves in Fig. 6. We use F_1 to denote the free energy of the solid phase and F_2 to denote that of the liquid phase. The condition of phase equilibrium has the form

$$F_2-F_1+p\Delta v=0, \tag{21}$$

where $\Delta v=v_2-v_1$ is the volume jump upon melting. At relatively small Δv, F_2 can be expanded in Δv:

$$F_2(v+\Delta v)=F_2(v_1)+\frac{\partial F_2(v_1)}{\partial v}\Delta v-U(v_1)-\frac{\partial U}{\partial v}\Delta v,$$

where $F_2(v_1)$ is the thermal component of the free energy of the liquid when $\Delta v=0$. It differs from the corresponding energy of the crystal by another value of the Debye temperature: $\Theta_2=\Theta_1-\Delta\Theta$, where $\Delta\Theta$ is the jump in the Debye temperature upon passage from the solid to the liquid. Therefore, if $\Delta\Theta/T\ll 1$, the relation $F_1(v)\cong F_2(v)$ holds on the high-temperature portion of the melting curve. Then $F_2-F_1\cong p\Delta v$, and condition (21) is satisfied. A more interesting consequence follows from the equality between the pressures of the phases $p_2=p_1$. On the solidification and melting curves we have $\Theta/T\ll 1$ and $D(\Theta/T)\cong 1$. As follows from Fig. 6, the contribution of the elastic component to the pressure can be neglected at high temperatures. Then the pressures of the phases are equal if

$$\frac{\Gamma_2+L_2/3}{1+L_2}=\frac{\Gamma_1+L_1/3}{1+L_1}. \tag{22}$$

Since $L_1,L_2\ll 1$ in the melting region, (22) reduces to the simpler condition

$$\Gamma_2+\frac{L_2}{3}=\Gamma_1+\frac{L_1}{3}. \tag{23}$$

As was shown above, Γ_1 and Γ_2 maintain constant values on the solidification and melting curves ($\Gamma_1=2.67$, $\Gamma_2=2.57$). Therefore, L_1 and L_2 should also remain constant. This is equivalent to Lindemann's hypothesis. Therefore, in the interpolation model considered here, this hypothesis follows from the condition of equality between the thermal components of the pressures of the phases during melting.

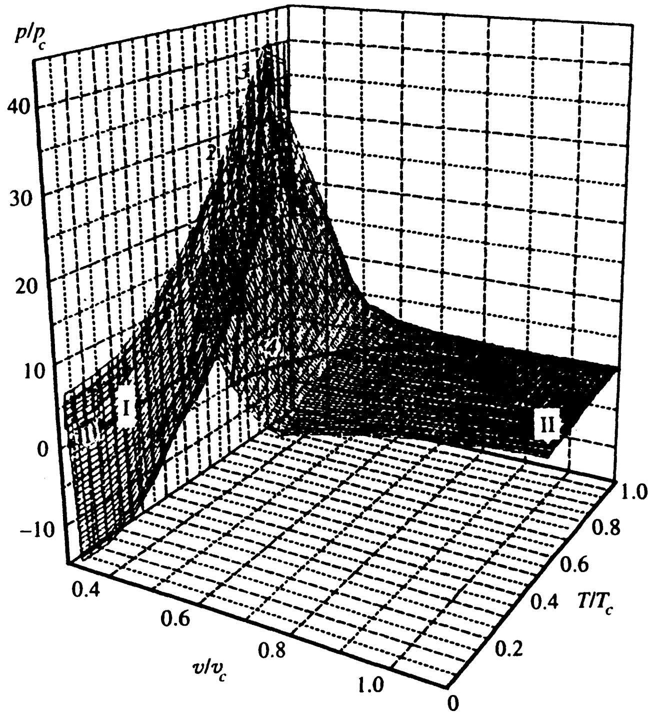

FIG. 7. $p(v,T)$ surfaces of neon: I—solid state; II—liquid and gas states; *1–4*—phase equilibrium curves.

The data obtained are sufficient for formulating a broad-range equation of state for Ne. To illustrate this, $p(v,T)$ surfaces have been constructed in the $v/v_c-T/T_c$ plane in Fig. 7. Surface I corresponds to the solid state. The cross sections formed by $T=$const planes form a family of isotherms, which monotonically decrease with increasing v. Surface II is the fluid surface, that exists at $T\geqslant T_t$. Its isotherms at $T<T_c$ have the van der Waals form. The solid and fluid surfaces do not intersect. Passage from one surface to the other occurs on freezing curve *2* and melting curve *3* at constant pressure. Sublimation curve *1* and vaporization curve *4* are also plotted in Fig. 7.

Thus, the generalized Debye approximation proposed here has made it possible to obtain a unified description of the passage of a substance from the solid crystalline state to the gaseous state, including the melting region. The function $\Theta_D(v)$ appearing in this approximation was found from tables of thermodynamic functions. Their approximation is thereby minimized. This makes it possible to calculate any thermodynamic quantity comparatively easily from Eqs. (13)–(19) with an accuracy corresponding to the accuracy of the data in Ref. 8. However, this does not exhaust the results following from the generalized approximation. The very possibility of using ordinary terms to describe the phases of a substance that are difficult to calculate opens up a new approach for investigating them. The interpretation of the reasons for the constancy of the Lindemann parameter in the melting region obtained in the present work is fairly convincing, but not the only illustration of the possibilities of such an approach.

This research was supported by the Russian Fund for Fundamental Research (project No. 94-02-03809).

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