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Volume change on melting for systems with inverse-power-law interactions

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We consider the change in volume on melting for systems interacting with a repulsive r^{-n} potential. Using the scaling property for inverse-power-law systems and the Clausius-Clapeyron equation, we show that the change in volume on melting must go to zero as $n \rightarrow D$, the dimensionality of the system. For $n \leq D$ finite excess thermodynamic properties can be defined by using a uniform neutralizing background. We produce a system with a uniform background as the limit of a classical two-component system and again find no change in volume on melting. Possible implications of these results for simulation studies of two-dimensional melting are discussed.

I. INTRODUCTION

Generally at the first-order melting transition there is a nonzero change $\Delta v = v_l - v_s$ in the molar volume of the coexisting fluid and solid phases, as well as the change Δs in molar entropy which manifests itself in the latent heat of melting.[1] However a nonzero Δv is by no means required for a first-order transition. The Clausius-Clapeyron equation[1] describes the rate of change of the melting temperature with pressure as

$$\frac{dT_m}{dP} = \frac{\Delta v}{\Delta s} \quad . \tag{1}$$

If $\Delta v = 0$, Eq. (1) shows that the melting curve has a maximum. Melting-point maxima have been observed experimentally for Cs (Ref. 2) and Rb (Ref. 3) and the Gaussian core model[4] offers a well-defined theoretical system which also has a melting-point maximum. In the latter system the potential of mean force becomes "softer" as the pressure or density is increased, and it seems likely that such an effect is operative also in the alkali metals.[5]

In this note we point out that the change in volume on melting also tends to zero for a system with a repulsive inverse-power-law pair potential r^{-n} in the limit $n \rightarrow D$ with D the dimensionality of the system. Unlike the previous examples, this is a consequence of the divergence of the pressure P because of the increasingly long-ranged nature of the potential in this limit. For $n \leq D$ finite excess pressure and other thermodynamic properties can be defined by considering a "uniform neutralizing background" (explained below). However the change in volume on melting again is zero if proper account of the background pressure is taken. Our treatment of the background may also help clarify some apparently anomalous properties of the frequently studied "one-component plasma" (OCP).[6]

For $D = 2$ it is possible that melting proceeds by two second-order transitions[7] with Δs as well as $\Delta v = 0$. We can shed no light on this interesting possibility but point out that even if the transition is first order with finite Δs, the change in volume must be zero for the above systems. This has some implications for computer simulations of two-dimensional (2D) melting which will be discussed at the end of this paper. Finally we note that any fundamental theory of first-order melting must also be capable of explaining the above examples of melting with no change in volume.

II. SCALING AND THE CLAUSIUS-CLAPEYRON EQUATION

Inverse-power-law systems have a useful scaling property[8,9] which arises because there is no temperature-independent characteristic length in the potential. The temperature $T \equiv \beta^{-1}$ (for notational simplicity we suppress the printing of Boltzmann's constant k_B) combines with the pair potential $u(r)$ in the partition function only in the combination

$$\beta u(r) = \beta\epsilon(\sigma/r)^n \quad . \tag{2}$$

Here r is the distance between a pair of particles and ϵ has dimensions of energy and σ length. In general for a two-parameter potential, the dimensionless *excess* thermodynamic properties (i.e., those which arise because of the interaction potential in excess of the ideal-gas contributions) are functions of two independent dimensionless state variables, the dimensionless reduced density $\rho\sigma^D$ and the reduced inverse temperature $\beta\epsilon$. Here the number density $\rho = N/V = v^{-1}$ with N the number of particles, V the volume, and v the molar volume. However, for the inverse-power-law potential, Eq. (2), we can define $\sigma_* = (\beta\epsilon)^{1/n}\sigma$ and Eq. (2) becomes $\beta u(r) = (\sigma_*/r)^n$. The temperature no longer appears explicitly and hence the dimensionless excess thermodynamic properties depend only on the single remaining variable[8,9]

$$\rho_* \equiv \rho\sigma_*^D = \rho\sigma^D(\beta\epsilon)^{D/n} \; . \tag{3}$$

(We now take σ and ϵ as units of length and energy so they will not appear explicitly in the rest of this paper.) Thus a single isotherm is sufficient to determine the entire equation of state. The melting curve in the entire T, ρ plane is described by some constant value of ρ_* and anywhere along the melting curve the dimensionless excess thermodynamic properties have the same value.[8,9]

We can exploit this scaling property in a novel way by considering its implications along with the Clausius-Clapeyron equation, Eq. (1). From scaling we know that the dimensionless compressibility factor $\beta P/\rho$ (the ideal-gas value can be added on trivially) is a function only of ρ_*. This must also be true for $\rho_*(\beta P/\rho) = \beta^{1+D/n}P$, using Eq. (3), and hence

$$P = T^{1+D/n} f(\rho_*) \tag{4}$$

for some function f. Along the melting curve ρ_* and $f(\rho_*)$ are constant and hence Eq. (4) can describe the variation of pressure with temperature along the melting curve. In particular, we have

$$\left(\frac{dP}{dT}\right)_{\rho_*} = \left(1+\frac{D}{n}\right)T^{D/n} f(\rho_*) = \left(1+\frac{D}{n}\right)\beta P \; . \tag{5}$$

Substituting Eq. (5) into Eq. (1) we find our basic result

$$\Delta s = \left(\frac{\rho^s-\rho^l}{\rho^l}\right)\left(1+\frac{D}{n}\right)\left(\frac{\beta P}{\rho^s}\right) \; , \tag{6}$$

where the terms grouped together by parentheses are all functions of the scaling variable. The change in molar entropy Δs is thus given by the product of the compressibility factor at melting times the fractional change in density times a factor $(1+D/n)$ which varies slowly with D and n. For $D=3$ the computer-simulation data of Hoover *et al.*[8] for inverse-power-law systems with $n=4$, 6, 9, 12, and ∞ are consistent with Eq. (6) to within their estimated error, as can be seen from Table I.

Let us now consider the n dependence of the various terms in Eq. (6). Computer-simulation data for $D=3$ (see Table I) are in agreement with the intuitive expectation that Δs is a weak (and, if anything, decreasing) function of n^{-1}. Since Δs measures the difference in order in the two phases and can be formally represented using only the singlet and pair distribution functions,[10] it must remain well defined and finite even in the limit $n \rightarrow D$ where the potential becomes long ranged. A proof of this physically obvious fact is given in the Appendix.

On the other hand $\beta P/\rho^s$ at melting is a rapidly increasing function of n^{-1}. This arises from two causes. First, if n is decreased at constant density, the melting temperature T_m decreases. This can be rationalized from Lindemann's law, since the softer potential permits larger vibrations at lower temperatures. From the data in Table I for $D=3$ this seems to be the major effect until $n=4$. Second, as the potential becomes longer ranged, more and more neighbors contribute positive terms to the pressure. The increase in P_m with decreasing n at constant density is also evident from the data in Table I. The resulting large increase in $\beta P/\rho^s$ requires a rapid decrease in the fractional change in density on melting $\Delta\rho/\rho^l$ since their product times the slowly varying factor $(1+D/n)$ equals the more or less constant Δs. Already for $n=4$ there is a fractional change in density on melting of only 0.5%.

The decrease in $\Delta\rho/\rho^l$ must continue even more dramatically as $n \rightarrow D$ where the long-ranged nature of the potential becomes dominant. Indeed from the

TABLE I. Data from Hoover *et al.* (Ref. 8) for $D=3$ and various values of n. The last three columns give the pressure and temperature (calculated by scaling) where the soft-sphere system melts at the same density $\rho_0 = 1.0408$ that the hard sphere ($n=\infty$) system melts, and the value of the scaling parameter ρ_*^s. Note that ρ_*^s must remain finite as $n \rightarrow D$ as long as there is a nonzero melting temperature for some fixed density.

n	Δs	$\Delta\rho/\rho^l$	$\beta P/\rho^s$	$P_m(\rho^s=\rho_0)$	$T_m(\rho^s=\rho_0)$	ρ_*^s
∞	1.16	0.103	11.28	a	a	1.04
12	0.90	0.038	18.96	6.78	0.578	1.19
9	0.84	0.030	22.66	8.58	0.435	1.37
6	0.75	0.013	39.10	9.08	0.223	2.20
4	0.80[b]	0.005	108.12	12.04	0.107	5.56

[a] Hard-sphere melting is independent of temperature so T_m and P_m separately are not well defined.

[b] Hoover *et al.* estimate an error of ±0.2 in Δs for this value of n.

virial equation[11]

$$\frac{\beta P}{\rho}=1-\frac{\beta\rho}{2D}\int d\vec{r}\,ru'(r)g(r) \tag{7}$$

we see on approximating the radial distribution function $g(r)$ by unity for large r that as n approaches D, $\beta P/\rho$ diverges as $(n-D)^{-1}$. Since Δs remains finite as this limit is taken, it follows from Eq. (6) that $\Delta\rho/\rho^l$ must tend to zero.

III. LONG-RANGED FORCES AND THE UNIFORM NEUTRALIZING BACKGROUND

For $n \leq D$ the pressure from Eq. (7) is infinite and we must proceed more carefully. The usual procedure[6,12–17] is to introduce a "uniform neutralizing background." As indicated below, the excess thermodynamic properties are then finite and obey scaling relations. However anomalous features appear unless proper attention is paid to the background. For example for $D=3$ the OCP with $n=1$ has negative pressure and isothermal compressibility at relatively low density if no account of the background pressure is taken.[6,12,13] Some workers have suggested the possibility of a phase transition because of these negative values.[14–16] Others[12,13] have treated the background as a degenerate (quantum) electron gas in order to describe a physical model appropriate for dense stellar interiors. Pollock and Hansen[17] pointed out that the change in volume on melting for the OCP depends explicitly on the nature of the background. Assuming a degenerate electron gas they estimated from their computer-simulation work for the $D=3$ OCP an extremely small fractional change in volume on melting of 0.03%.[17]

While this treatment of the background is appropriate for the physical application made by Pollock and Hansen,[17] we believe the simplest and most consistent way of introducing a uniform background in a *classical* treatment is to consider it as a well-defined limit (to be described below) of classical two-component system. Using this classical background, the *total* pressure and compressibility are always non-negative, and there can be no change in volume on melting, in agreement with the limiting value as $n \to D$ from above.

To describe a system with $n \leq D$ let us first imagine a two-component system made up N_2 "background particles," each with charge e_2, and the physically relevant component of N_1 "ions, "each with charge e_1. To avoid collapse in this classical system as oppositely charged particles approach close to one another, the ions have a hard core of radius R_1 (which will later tend to zero).[18] The interaction potential $u_{12}(r)$ is thus infinite if $r<R_1$ while $u_{11}(r)$ is infinite for $r<2R_1$. Otherwise the potential is

$$u_{ij}(r)=e_ie_j/r^n \ . \tag{8}$$

We require always that the system is electrically neutral:

$$N_1e_1+N_2e_2=\rho_1e_1+\rho_2e_2=0 \ . \tag{9}$$

The total pressure is given by the two-component version of Eq. (7)[19]

$$\beta P=\rho_1+\rho_2-\frac{\beta}{2D}\sum_{i,j=1,2}\rho_i\rho_j\int d\vec{r}\,ru_{ij}'(r)g_{ij}(r) \ . \tag{10}$$

The first terms on the right are the ideal-gas contributions to the pressure and $g_{ij}(r)$ is the radial distribution function between species i and j.

We now take the "dense-point limit" of Waisman and Lebowitz,[20] with $\rho_2 \to \infty$, $e_2 \to 0$ such that the charge density ρ_2e_2 remains constant, satisfying Eq. (9). Having sufficiently delocalized the background charge distribution by first taking the dense-point limit we can now let the hard-core radius $R_1 \to 0$ without fear of collapse. Correlations involving the background particles should vanish since e_2 is infinitesimal and we expect

$$g_{12}=g_{21}=g_{22}=1 \tag{11}$$

for all $r>0$. As is appropriate for a uniform background, there are no correlations involving the background particles.[21] Thus a density fluctuation $\delta\rho_1(k)$ for a nonzero wave vector is unaffected by the background. However a $k=0$ fluctuation, i.e., a finite change in charge density $\delta\rho_1e_1$ requires a corresponding change in background charge density $\delta(\rho_2e_2)$ so that the neutrality condition, Eq. (9), continues to hold.

The above assertion about the behavior of the $g_{ij}(r)$ as the various limits are taken holds true exactly for the mean spherical model solved by Waisman and Lebowitz[20] and seems physically very plausible in general. We assume in the following that Eq. (11) is satisfied; indeed this could be taken as a necessary requirement for a uniform background in a two-component system.[21] However, it is only in the dense-point limit that such correlation functions could arise physically as the limit of a classical two-component system.

Using Eq. (8), Eq. (10) can be written explicitly as

$$\beta P=\rho_1+\rho_2+\frac{\beta n}{2D}\sum_{i,j=1,2}\rho_ie_i\rho_je_j\int d\vec{r}\,r^{-n}g_{ij}(r) \ . \tag{12}$$

In the dense-point limit, using Eqs. (11) and (9), we find from Eq. (12)

$$\beta P=\rho_1+\rho_2+\frac{\beta n}{2D}(\rho_1e_1)^2\int d\vec{r}\,r^{-n}[g(r)-1] \ , \tag{13}$$

where $g(r) \equiv g_{11}(r)$. Thus the standard "background subtraction" with $g(r)$ in Eq. (7) replaced by $[g(r)-1]$ in Eq. (13) arises naturally from the appropriate limit of the two-component formula, Eq. (10).

The excess pressure in Eq. (13) can be either positive or negative. However the total pressure includes the ideal-gas contribution ρ_2 from the background which becomes arbitrarily large in the dense-point limit. Similarly the excess compressibility can be negative but the total compressibility $k_T = -(1/V) \times (\partial V/\partial P)_T$ is zero because of the diverging background pressure. Most discussions of the OCP have not considered these background contributions.

Since the ions are coupled to the background by neutrality only for a $k=0$ fluctuation one can define an "effective compressibility" relevant for small (but nonzero) wave-vector density fluctuations $\delta\rho_1(k)$ by calculating $(\partial P/\partial V)_T$ from Eq. (13) but ignoring the background contribution. It is this "effective compressibility" that is frequently discussed in the OCP literature[12–17] and this can indeed be negative. However, as pointed out by Viellefosse and Hansen[22] this does not imply an instability in the OCP. A density fluctuation $\delta\rho_1(k)(k \neq 0)$ creates a local charge fluctuation $e_1\delta\rho_1(k)$ which is not screened by the static background. The resulting cost in energy from Poisson's equation is proportional to k^{-2}, so small k density fluctuations are strongly suppressed.[22] As we have argued above, the same is true for a $k=0$ density change if one takes proper account of the background pressure.

A system with this classical background can support no change in volume on melting. Indeed if we require equal pressures from neutral systems with fluid densities ρ_{1l} and ρ_{2l} and solid densities ρ_{1s} and ρ_{2s} we have from Eq. (13)

$$\rho_{1l} + \rho_{2l} + \beta P_{\text{ex}}(\rho_{1l}) = \rho_{1s} + \rho_{2s} + \beta P_{\text{ex}}(\rho_{1s}) \ , \qquad (14)$$

where P_{ex} is the excess pressure in Eq. (13). Charge neutrality in each phase requires from Eq. (9)

$$e_1\Delta\rho_1 + e_2\Delta\rho_2 = 0 \ , \qquad (15)$$

where $\Delta\rho_1 = \rho_{1s} - \rho_{1l}$. Multiplying Eq. (14) by e_2 we find on taking the dense-point limit with $e_2 \to 0$ that all terms vanish except those involving ρ_2 explicitly and we have, using Eq. (15),

$$0 = \Delta\rho_2 e_2 = \Delta\rho_1 e_1 \ . \qquad (16)$$

Thus $\Delta\rho_1 = 0$ as stated above.

Having established that the background permits no change in volume on melting we can for most practical purposes ignore it. It is easy to show that the excess thermodynamic properties again obey scaling with $\rho_* = \rho_1\beta^{D/n}$ just as in Eq. (3). In computer simulations of melting, the background has no effect on the dynamics and anywhere along an isochore the background contribution to βP has the same constant (but formally infinite) value. Indications of melting will show up in the *excess* thermodynamic properties as well as from the usual structural and dynamic indicators.

IV. FINAL REMARKS

Although we have considered only repulsive r^{-n} potentials we expect that the same qualitative features will hold for most other smooth repulsive potentials which give a positive change in volume on melting. An effective exponent n_{eff} can be defined in a number of ways, e.g.,

$$n_{\text{eff}}(r) = -\frac{d \ln u(r)}{d \ln r} \qquad (17)$$

evaluated at the nearest-neighbor distance. [Equation (17) gives the exponent n for any r for an inverse-power-law potential.] We expect $\Delta\rho/\rho^l$ to be a decreasing function of n_{eff}. In the limit that the potential becomes long ranged with $\beta P/\rho$ diverging, $\Delta\rho$ must again be zero. Most realistic potentials have attractive as well as repulsive regions. The effect of attractions can often be accurately treated by a simple mean-field theory.[23] This approach predicts a larger change in volume than that arising from the repulsive interactions alone.

Currently there is much debate about the interpretation of computer-simulation data for the melting of a 2D Lennard-Jones solid.[24] Along an isochore there are two breaks at temperatures T_1 and T_2 in the thermodynamic and other properties, but it is difficult to tell whether these signify two second-order transitions with an intermediate hexatic phase as suggested by Halperin and Nelson[7] (HN), or an ordinary first-order transition with a two-phase region between T_1 and T_2.[25]

The results of this paper suggest that it might be profitable to study inverse-power-law potentials for small values of n. If the transition is first order then the two-phase region will be of very narrow or vanishing width. However, if melting proceeds by two second-order transitions, none of the arguments in this paper constrain the width of the hexatic phase in any way. The finding of a wide intermediate region would strongly favor the HN theory.

Indeed it seems plausible that the softer interactions for small n might enhance the possibility of a second-order melting transition. The system is far from the very anharmonic hard-disk limit where geometric-packing considerations dominate (and the melting transition is believed to be first order[25,26]). The basic HN picture of phonons interacting with bound dislocation pairs seems more appropriate for these soft potentials. There have been several simulations of the $n=1$ OCP[27] and we have begun a study of a cutoff version of the $n=3$ system.[28] These systems may offer a favorable testing ground for the possibility of a second-order melting transition in two dimensions.

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APPENDIX

It is perhaps worthwhile to indicate more explicitly why Δs must remain finite as $n \rightarrow D$. It is trivial to show that the entropy of any classical system is less than the (finite) ideal-gas entropy. Equation (6) shows that $s^l > s^s$ so the only way to obtain an infinite Δs is for $s^s \rightarrow -\infty$. Since the third law does not apply to classical systems this cannot be immediately ruled out. However we can make use of the variational principle[10] for the excess Helmholtz free energy $(\beta A_{\rm ex}/N)$ to show that s^s is finite.

The excess free energy must be less than that of a constrained "single-occupancy" system where each particle is confined to its $T=0$ Wigner-Seitz cell. This in turn is less than the free energy obtained by approximating the N-particle distribution by a product of single-particle distribution functions. We have, as discussed in detail by Hill,[29]

$$\frac{\beta A_{\rm ex}}{N} - \ln\rho + 1 \leq \sum_{j=2}^{N} \int_{v_1}\int_{v_j} \beta u(r_{ij})\phi(\vec{r}_1)\phi(\vec{r}_j)\,d\vec{r}_1\,d\vec{r}_j - \int_{v}\phi(\vec{r})\ln\phi(\vec{r})\,d\vec{r} \ , \tag{A1}$$

where the first term gives the approximate energy and the second the entropy. The integrations are restricted to Wigner-Seitz cells of volume $v=\rho^{-1}$ and the center of the ith cell is chosen as the origin for $\vec{r}_i$. Here $\phi(\vec{r})$ is a trial single-particle distribution function, which we can choose arbitrarily and still obtain an upper bound to the true free energy. To obtain a lower bound to the entropy at melting

$$s_{\rm ex}^{s} = \frac{\beta E_{\rm ex}}{N} - \frac{\beta A_{\rm ex}}{N} \tag{A2}$$

we substitute into Eq. (A2) a lower bound $\beta E_{\rm ex}^{0}/N$ to the energy, where $E_{\rm ex}^{0}$ is the $T=0$ potential energy, and an upper bound to $\beta A_{\rm ex}/N$ obtained from Eq. (A1). Combining the energy terms we have

$$s_{\rm ex}^{s} \geq \ln\rho - 1 + \sum_{j=2}^{N} \int_{v_1}\int_{v_j} \beta\Delta u(r_{1j})\phi(\vec{r}_1)\phi(\vec{r}_j)\,d\vec{r}_1\,d\vec{r}_j + \int_{v}\phi(\vec{r})\ln\phi(\vec{r})\,d\vec{r} \ . \tag{A3}$$

We have noted that $\phi(\vec{r})$ is normalized and defined

$$\Delta u(r_{1j}) \equiv u(r_{1j}^{0}) - u(r_{1j}) \ , \tag{A4}$$

where $u(r_{1j}^{0})$ is the (constant) value of the pair potential energy when both particles are on their $T=0$ lattice sites with separation r_{1j}^{0}. Since Δu rather than u appears in Eq. (A3) we obtain a finite result even if $n \rightarrow D$ as long as we choose a $\phi(\vec{r})$ which prevents particles in nearest-neighbor cells from approaching arbitrarily close to one another and themselves producing an infinite energy. For example the constant probability $\phi(\vec{r}) = 1/v_s'$ for $\vec{r}$ contained with a smaller cell of volume $v_s' < v$ [and $\phi(\vec{r})=0$ otherwise] gives a finite lower bound to $s_{\rm ex}^{s}$.

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