Classical nucleation theory of the one-component plasma

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We investigate the crystallization rate of a one-component plasma (OCP) in the context of classical nucleation theory. From our derivation of the free energy of an arbitrary distribution of solid clusters embedded in a liquid phase, we derive the steady-state nucleation rate of an OCP as a function of the Coulomb coupling parameter Γ. Our result for the rate is in accordance with recent molecular dynamics simulations, but it is greater than that of previous analytical estimates by many orders of magnitude. Further molecular dynamics simulations of the nucleation rate of a supercooled liquid OCP for several values of Γ would clarify the physics of this process.

I. INTRODUCTION

The one-component plasma (OCP) consists of a single species of N charged ions of mass m in a volume V in a uniform neutralizing electron background. This idealized system is a model for astrophysical settings such as neutron star crusts and white dwarf interiors. The dimensionless coupling parameter

$$\Gamma = \frac{(Ze)^2}{a k_B T} \qquad (1)$$

characterizes the state of the OCP, where Ze is the ion's charge, $a=(3/4\pi n_I)^{1/3}$ is the mean distance between ions at number density $n_I = N/V$, and T is the temperature. We consider the ions to be classical with a de Broglie wavelength $\lambda = (h^2/2\pi m k_B T)^{1/2} \ll a$. For $\Gamma \ll 1$, the Coulomb coupling is weak and the ions behave like an ideal Maxwell-Boltzmann gas. For $\Gamma \gg 1$, the Coulomb coupling is large and the system is in the liquid phase until $\Gamma > \Gamma_m \approx 175$ [1,2], when the ions undergo a first-order phase transition into a periodic lattice with a body-centered-cubic configuration.

The Helmholtz free energy of the OCP in the liquid phase is written as $F = F^{id} + F^{ex}$, where the ideal part $F^{id} = Nk_B T[\ln(n_I \lambda^3) - 1]$, and interactions are incorporated into the "excess" part $F^{ex} = -k_B T \ln(Z_N/V^N)$, where Z_N is the configuration integral [3]. In the noninteracting limit, $Z_N \to V^N$ and $F^{ex} \to 0$. Simulations of the liquid OCP have generated accurate fitting formulas for the reduced excess free energy $f^{ex}(\Gamma) = F^{ex}/Nk_B T$ (e.g., [4]), and calculations of the solid-state free energy (e.g., [5]) allow for the determination of Γ_m.

Crystallization of an OCP occurs via nucleation, the process by which small crystals form via localized fluctuations in the liquid and grow via the accretion of surrounding ions (see, e.g., [6]). Empirically, most pure liquids can be supercooled (in our case $\Gamma > \Gamma_m$) without solidifying, which implies that a kinetic barrier to nucleation exists (e.g., [7]). Monte Carlo [8,9] and molecular dynamics [10] simulations demonstrate that such a kinetic barrier exists in an OCP as well. Localized fluctuations of sufficient amplitude can generate crystals large enough to overcome the kinetic barrier,

and hence they are thermodynamically stable. Such crystals subsequently grow and facilitate the phase transition.

The rate at which astrophysical solids grow can affect the degree of chemical phase separation during the solidification of both white dwarf interiors ([11], and references therein) and neutron star crusts [12], the existence of an amorphous glassy state in white dwarf interiors [13,14], and the prevalence of defects and impurities in neutron star crusts [15,16]. Furthermore, the time at which the latent heat of crystallization is released within a white dwarf affects its observed cooling rate (e.g., [17]). Unfortunately, the nucleation rate of an OCP is not well understood. Several authors [14,15,18,19] assumed that classical nucleation theory [6] applied to the OCP, but the accuracy of their assumptions was untested. Daligault [10] was the first to directly study nucleation of an OCP using molecular dynamics simulations. Although his results were qualitatively consistent with classical nucleation theory, it was unclear whether or not the results agreed quantitatively. In this paper, we reexamine the nucleation of an OCP in the context of classical nucleation theory, but with a different approach to the statistical physics of an ensemble of solid clusters embedded in a liquid phase. We find a steady-state nucleation rate as a function of Γ in quantitative accordance with those of [10].

We begin in Sec. II by reviewing what is known about the excess free energies for multicomponent plasmas and then deriving the minimum reversible work of cluster formation and the equilibrium distribution of cluster sizes. We use these results to deduce the nucleation rate in Sec. III, and we compare this rate, along with that used in previous work, to the results of [10] in Sec. IV. We close in Sec. V by discussing our results and suggesting future numerical experiments and analytic improvements to this work.

II. MINIMUM REVERSIBLE WORK OF CLUSTER FORMATION

We consider a classical OCP consisting of N_I ions of mass m in a uniform neutralizing background of electrons at $n_e = Z n_I$. Motivated by the typical astrophysical context, we also assume that degenerate electrons supply all of the pressure so

that n_e is constant during any phase change and set by charge neutrality. However, before discussing how we treat solid clusters embedded in a liquid OCP, it is important to clarify what is known about mixtures of ions of different charges.

Hansen *et al.* [20] first showed that the excess Helmholtz free energy in a mixture of ions of charges Z_i and number N_i in a volume V is simply given by the sums of the excess free energies of each separate species at the same electron density:

$$F^{ex} = k_B T \sum_i N_i f^{ex}(\Gamma_i). \qquad (2)$$

Here $\Gamma_i = Z_i^{5/3} \Gamma_e$ and $\Gamma_e = e^2/a_e k_B T$, where $a_e = (3/4\pi n_e)^{1/3}$ is the mean distance between electrons, and all $\Gamma_i > 1$. Recent simulations [4] have shown that the deviations from this simple linear mixing rule are less than 0.05%. Hence, to an excellent approximation, the free energy of a mixture of liquid ions is simply

$$\frac{F_{total}}{k_B T} = \sum_i N_i \left[\ln\left(\frac{N_i \lambda_i^3}{V} \right) - 1 \right] + \sum_i N_i f^{ex}(\Gamma_i). \qquad (3)$$

Later authors chose to write this equation differently by using the fits f_l of the total Helmholtz free energies of each pure state and then mixing them. This results in the more often seen equation

$$\frac{F_{total}}{k_B T} = \sum_i N_i f_l(\Gamma_e Z_i^{5/3}) + \sum_i N_i \ln\left(\frac{Z_i N_i}{\langle Z \rangle N_{total}} \right), \qquad (4)$$

where $\langle Z \rangle$ is the average charge. The last term is minus the entropy of mixing (see e.g., [20,21] and the Appendix), although it originates simply from charge conservation at a fixed n_e. This term will appear in our derivation of the minimum reversible work of cluster formation.

This knowledge of mixing at fixed n_e motivates our rederivation of the minimum reversible work of cluster formation, and thereby the equilibrium cluster distribution, from the total Helmholtz free energy F_{total} of an arbitrary distribution of clusters. Following Frenkel [22] (see also [23,24]), we consider clusters that contain different numbers of ions to be distinct species, and we consider all clusters that contain the same number of ions to be indistinguishable. The latter is an approximation because such clusters could have different morphologies. Thus we assume that, for each cluster size, there exists a unique configuration of ions for which the total surface free energy is a minimum, that this configuration is spherical, and that all clusters of this size conform to this configuration.

The total Helmholtz free energy of the system is

$$\frac{F_{total}}{k_B T} = N_1 f_l + \sum_{i=2}^{\infty} N_i(i f_s + i^{2/3}\alpha)$$

$$+ \sum_{i=1}^{\infty} N_i \left[\ln\left(\frac{N_i}{N_{total}} \right) + \ln\left(\frac{Z_i}{\langle Z \rangle} \right) \right], \qquad (5)$$

where $f_l k_B T$ and $f_s k_B T$ are the free energies of one ion in the pure liquid and pure solid state, respectively, $\alpha k_B T$ is proportional to the surface free energy of an ion at the boundary of

a cluster, N_i and $Z_i = iZ$ are the number and charge, respectively, of clusters (including liquid monomers) that consist of i ions,

$$N_{total} = \sum_{i=1}^{\infty} N_i \qquad (6)$$

is the total number of clusters (including monomers), and

$$\langle Z \rangle = \sum_{i=1}^{\infty} Z_i \frac{N_i}{N_{total}} \qquad (7)$$

is the average charge of the clusters. Equation (5) is subject to the constraint

$$N_I = \sum_{i=1}^{\infty} i N_i. \qquad (8)$$

From Eq. (5), the chemical potentials of monomers and clusters are

$$\frac{\mu_1}{k_B T} = f_l + \ln\left(\frac{Z_1 N_1}{\langle Z \rangle N_{total}} \right) + 1 - \frac{Z_1}{\langle Z \rangle}, \qquad (9)$$

$$\frac{\mu_i}{k_B T} = i f_s + i^{2/3}\alpha + \ln\left(\frac{Z_i N_i}{\langle Z \rangle N_{total}} \right) + 1 - \frac{Z_i}{\langle Z \rangle}, \qquad (10)$$

respectively. Equation (9), the chemical potential of a liquid monomer, is identical to that of [25]. According to classical nucleation theory, clusters grow or shrink by the addition or subtraction of single monomers. Thus in chemical equilibrium $\mu_i = \mu_{i-1} + \mu_1$, and so

$$\mu_i = i\mu_1 \qquad (11)$$

by iteration. From Eqs. (9)–(11) and noting that $\langle Z \rangle N_{total} = Z N_I$, the equilibrium distribution of clusters is

$$\frac{i N_i}{N_I} = \left(\frac{N_1}{N_I} \right)^i \exp\{ -[i(f_s - f_l - 1) + i^{2/3}\alpha + 1] \}, \qquad (12)$$

which is in the form of the law of mass action. The monomer concentration, and hence the concentration of all clusters, is found by invoking the constraint of Eq. (8). Alternatively, Eq. (12) can be written in terms of the Boltzmann distribution $P(i) = \exp(-\Delta f_i)$, where $P(i) = i N_i/N_I$ is the probability that an ion picked at random is a member of a cluster of size i and

$$\Delta f_i = \begin{cases} -\ln(N_1/N_I), & i = 1, \\ i[f_s - f_l - 1 - \ln(N_1/N_I)] + i^{2/3}\alpha + 1, & i \geq 2, \end{cases} \qquad (13)$$

is the minimum reversible work divided by $k_B T$ of forming a cluster of size i. Demanding that $P(i)$ be normalized then yields N_1/N_I.

III. STEADY-STATE NUCLEATION RATE

We follow the treatment of classical nucleation theory by Kelton [6] and derive the steady-state cluster nucleation rate.

We consider an OCP at fixed Γ that is entirely in the liquid state at time $t=0$ and calculate the rate of cluster formation for $t>0$, when $N_1 \approx N_I$, and so

$$\Delta f_i = i(f_s - f_l - 1) + i^{2/3}\alpha + 1, \qquad (14)$$

from Eq. (13). We use the fit of [1] for $f_l(\Gamma)$, where we take the coefficients derived from the simulation data of [26], and we use the fit of [5] for $f_s(\Gamma)$. For sufficiently large Γ, $(f_s - f_l - 1) < 0$. The fit of f_s given by Eqs. (10) and (15) of [5] is valid only for $170 \leq \Gamma \leq 2000$, and the above inequality is easily satisfied for the entire range of Γ in which the fits are applicable. The minimum Γ above which $(f_s - f_l - 1) < 0$ is unknown. Since $\alpha > 0$, there exists a critical size

$$i_* = \left(\frac{2\alpha/3}{1 + f_l - f_s}\right)^3 \qquad (15)$$

such that the reversible work of cluster formation, Δf_i, is a maximum. Cluster growth is energetically favorable for clusters of size $i > i_*$ and unfavorable for clusters of size $i < i_*$. Equation (14) can now be written in the convenient form

$$\Delta f_i = \frac{(2\alpha/3)^3}{2(1 + f_l - f_s)^2}\left[3\left(\frac{i}{i_*}\right)^{2/3} - 2\left(\frac{i}{i_*}\right)\right] + 1. \qquad (16)$$

In classical nucleation theory, crystallization occurs via the formation and subsequent growth of critical clusters of size i_*. Clusters of size $i < i_*$ are formed via equilibrium fluctuations and are presumed to be transient. Equation (12) gives their distribution. Clusters of size $i > i_*$ are presumed to be stable. Such clusters only grow with time. The formation of stable clusters is therefore a two-step process: a stable cluster forms when (a) equilibrium fluctuations in the liquid phase generate a transient cluster of size i_* and (b) that cluster accretes an additional monomer. Therefore, the steady-state nucleation rate J, defined to be the number of solid clusters formed per second, is roughly equal to the number of critical clusters of size i_* times the rate at which a monomer attaches to a critical cluster. Kelton [6] performs a more thorough derivation of the nucleation rate and finds

$$J(N_I, \Gamma) = \frac{24 D i_*^{2/3}}{a^2} N_I \left(\frac{\Delta f_{i_*}}{3\pi i_*^2}\right)^{1/2} \exp(-\Delta f_{i_*}), \qquad (17)$$

where D is the diffusion coefficient and $(\Delta f_{i_*}/3\pi i_*^2)^{1/2}$ is the Zeldovich factor. We use Eq. (17) for the steady-state nucleation rate of an OCP. For D, we use the classical OCP diffusion coefficient [27]

$$D = 2.95\omega_p a^2 \Gamma^{-1.34}, \qquad (18)$$

where $\omega_p = [4\pi n_1 (Ze)^2/m]^{1/2}$ is the ion plasma frequency. Recent calculations have verified the accuracy of this expression for $\Gamma \lesssim \Gamma_m$ [28,29] as well as for $\Gamma \gtrsim \Gamma_m$ [30]. For the normalized surface free energy α, we follow [15] and [18] and set

$$\alpha = 4\pi\left(\frac{3}{4\pi}\right)^{2/3}\beta\lambda\frac{k_B T_m}{k_B T} = (36\pi)^{1/3}\beta\lambda\left(\frac{\Gamma}{\Gamma_m}\right), \qquad (19)$$

where $\beta \approx 0.5$ is the ratio of the interfacial energy and heat of fusion empirically derived from liquid metal experiments
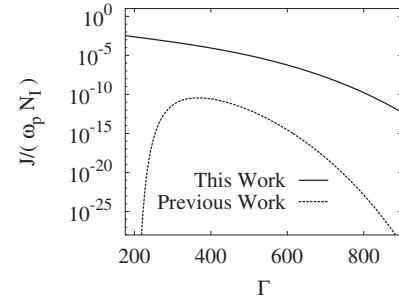

FIG. 1. Steady-state nucleation rate per ion as a function of Γ. "This Work" is the rate calculated from our model, and "Previous Work" is the rate calculated using the models of previous authors. Our calculations predict that the nucleation rate of a one-component plasma is $> 10^5$ times higher than previously assumed for all values of Γ.

(e.g., [31,32]) and $\lambda k_B T_m$ is the enthalpy of melting per ion. For this work, we set $\lambda = 0.77$ [5,33].

IV. COMPARISON TO PREVIOUS WORK

Several authors have investigated the homogeneous nucleation rate of an OCP [14,15,18,19]. However, in deriving the minimum reversible work of cluster formation Δf_i, they assumed that the entropy of mixing is simply that of an ideal gas; i.e., the term $\ln(Z_i/\langle Z\rangle)$ in Eq. (5) was omitted. Consequently, they find $\Delta f_i = i(f_s - f_l) + i^{2/3}\alpha$, and Eqs. (15) and (16) become

$$i_* = \left(\frac{2\alpha/3}{f_l - f_s}\right)^3, \qquad (20)$$

$$\Delta f_i = \frac{(2\alpha/3)^3}{2(f_l - f_s)^2}\left[3\left(\frac{i}{i_*}\right)^{2/3} - 2\left(\frac{i}{i_*}\right)\right]. \qquad (21)$$

We demonstrate below that previous calculations result in a nucleation rate that is orders of magnitude lower that the rates deduced from simulations, whereas our model predicts a rate closer to that found numerically. The steady-state nucleation rate per ion as a function of Γ for both models is shown in Fig. 1.

Presently, crystallization of an OCP is studied only via numerical simulations. Although there are many calculations of the free-energy difference of the pure liquid and pure solid states as a function of Γ, few have investigated the onset of crystallization. Ogata [8] and DeWitt et al. [9] studied nucleation in an OCP using Monte Carlo simulations. Unfortunately, time is not a variable in Monte Carlo simulations, so that the rate of nucleation is difficult to calculate (although see [34]).

Recently, Daligault [10] performed molecular dynamics simulations to investigate the kinetics of nucleation in a supercooled OCP. Using $N_I = 4394$ ions, he calculated the total number of solid nuclei as a function of time for $\Gamma = 400$ and 300. In the $\Gamma = 400$ calculation (see Fig. 4 of [10]), following a short period of transient nucleation, there is a significant period of time during which steady-state nucleation takes place, as evidenced by the nearly constant slope in the plot of

TABLE I. Steady-state nucleation rates and critical cluster sizes for $N_I = 4394$: "J_{MD}" is the inferred nucleation rate from the simulations of [10], "(this work)" denotes a quantity calculated from our model, and "(previous)" denotes a quantity calculated using the model of previous authors.

Γ	J_{MD}/ω_p	J/ω_p (this work)	J/ω_p (previous)	i_* (this work)	i_* (previous)
400	≈ 0.2	0.4	1×10^{-7}	4	51
300	~ 0.1	2	2×10^{-8}	3	91

the number of clusters as a function of time. In the $\Gamma = 300$ calculation (see Fig. 8 of [10]), there is no obvious period of time during which steady-state nucleation occurs. To be consistent with our estimate from the previous calculation, we estimate J_{MD}, the steady-state nucleation rate deduced from molecular dynamics simulations, from the period of time during which the nucleation rate is nearly constant, which occurs just before nucleation saturates. In Table I, we list the inferred values of J_{MD} as well as the nucleation rates calculated using both Eqs. (15) and (16) of our model and Eqs. (20) and (21) from previous models. Our model predicts nucleation rates that are in reasonable agreement with the results of Daligault [10], whereas the predictions of previous models are far too low.

V. DISCUSSION

Through an improved treatment of the statistical physics of the distribution of solid clusters in the liquid phase, we have developed a model of steady-state nucleation in an OCP, and the results of our model are in accordance with those of the time-dependent molecular dynamics simulations of [10]. Our results suggest that crystallization in an OCP occurs at a rate that is more than five orders of magnitude higher than previously assumed.

Nucleation in an OCP differs from nucleation in many other media because of two unique features of an OCP: (a) The degenerate electron background, which supplies the pressure, is unaffected during the phase transition. Consequently, both pressure and volume remain constant. Although the complete noninteraction of the electrons is an approximation of the OCP model, the role electrons play in more realistic models is negligible [1]. (b) The binding of ions of like charge in a crystal is weak. This is reflected in the difference of the liquid and solid free energies. Indeed, this difference does not exceed $k_B T$ (i.e., $f_l - f_s$ does not exceed 1) until $\Gamma/\Gamma_m = T_m/T \gtrsim 2.8$. The combination of these two features has interesting implications for the crystallization of an OCP. In particular, nucleation in an OCP is driven to a significant degree simply by the increase in entropy resulting from cluster formation: the change in the total energy of a supercooled OCP when a cluster forms is small relative to the increase in the entropy. In fact, Eq. (14) implies that a population of solid clusters will exist at a given time for a range of $\Gamma < \Gamma_m$ because the entropy of an ensemble of clusters embedded in a liquid OCP is higher than that of the pure liquid phase. This may be related to the caging effect observed in molecular dynamics simulations of an OCP in the liquid phase [28,35]. Furthermore, we note that a reanalysis of the entropy of mixing term and thereby the classical nucleation rate may be needed for other supercooled liquids.

There are a few important issues that we have not addressed. The surface free energy of a solid cluster in a Coulomb liquid is not known. The expression for the surface free-energy contribution used in this work is only our best estimate, but we cannot be confident that the numerical coefficient in Eq. (19) is accurate. This issue requires further investigation via molecular dynamics simulations. Some of the assumptions we made in Sec. II may not be valid. We assume that solid clusters grow or shrink by the addition or subtraction of liquid monomers, but the simulations of Daligault [10] imply both that two solid clusters can fuse together and that a solid cluster can fission into two or more clusters. Furthermore, we assume that all clusters of a given size have the same morphology and are thus indistinguishable, but the simulations of Daligault [10] show that two clusters of the same size can in fact have different morphologies. We currently do not know what effects, if any, a violation of either of these assumptions would have on our results.

Further molecular dynamics simulations are imperative to understand nucleation in an OCP. It is evident from Eqs. (17)–(19) that the steady-state nucleation rate may be written as

$$J = N_I \omega_p F(\Gamma), \qquad (22)$$

where $F(\Gamma)$ is a function only of Γ. Numerically confirming that the nucleation rate scales linearly with N_I and ω_p should be simple. One could then deduce $F(\Gamma)$, and hence J, by measuring the nucleation rate for several different values of Γ. Calculations for $\Gamma_m \lesssim \Gamma \lesssim 210$ would be particularly useful. Figure 1 shows that there is an enormous difference between our results and those of previous authors: our model predicts that some clusters should form rather quickly, whereas other models predict that clusters essentially never form.

APPENDIX: ENTROPY OF MIXING IN A ONE-COMPONENT PLASMA

We follow the treatment of [36] and derive the ideal entropy of mixing of clusters in an OCP. The entropy of an ideal system of N_i indistinguishable particles of mass m_i at temperature T and within a volume V is

$$\frac{S_i(V)}{k_B} = N_i\left[\ln\left(\frac{V}{N_i}\right) + \frac{5}{2} + \frac{3}{2}\ln\left(\frac{2\pi m_i k_B T}{h^2}\right)\right]. \quad \text{(A1)}$$

The ideal entropy of mixing is thus

$$\Delta S_{\text{mixing}} = \sum_{i=1}^{\infty} [S_i(V_{\text{total}}) - S_i(V_i)], \quad \text{(A2)}$$

where

$$V_i = \frac{Z_i N_i}{n_e} \quad \text{(A3)}$$

is the fractional volume occupied by the N_i clusters of charge Z_i and

$$V_{\text{total}} = \sum_{i=1}^{\infty} V_i = \frac{\langle Z\rangle N_{\text{total}}}{n_e} \quad \text{(A4)}$$

is the total volume of the system. The V_i's are set by demanding both local charge neutrality and that n_e be constant throughout the medium. It follows that

$$\frac{\Delta S_{\text{mixing}}}{k_B} = \sum_{i=1}^{\infty} N_i \ln\left(\frac{V_{\text{total}}}{V_i}\right) \quad \text{(A5)}$$

and hence

$$\frac{\Delta S_{\text{mixing}}}{k_B} = -\sum_{i=1}^{\infty} N_i \ln\left(\frac{Z_i N_i}{\langle Z\rangle N_{\text{total}}}\right). \quad \text{(A6)}$$

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