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Large-Scale Molecular Dynamics Simulation of Coulomb Clusters: A Finite-Temperature Analysis

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Thermal behavior of Coulomb clusters in a three dimensional confining potential is investigated by molecular dynamics simulations for system sizes of 1,000 to 20,288 ions. The specific heat of the system of shell-structured 20,000 ions is peaked almost at the same temperature as the system of bcc-structured 20,288 ions with much sharper structure for the latter. The diffusion coefficient and the peak to valley ratio of the two-dimensional pair distribution function on the outermost shell are obtained both as a function of temperature. The rotational movement of each shell in the system of 10^4 ions is observed.

1. INTRODUCTION

The laser cooled one-component plasma (Coulomb cluster) in *Penning-Malmberg* trap forms a crystal whose structure only depends on the number of ions. In experiments with small number of ions, concentric shell structures have been observed[1, 2]. In systems with more than $(1.1 \sim 1.5) \times 10^4$ ions, the formation of bcc crystals in the interior has been observed in molecular dynamics simulations[3].

Generally, the melting temperature of a cluster is lower than that of bulk material. Schiffer has suggested that the melting point of shell-structured clusters ($N < 10^4$) depends on the number of ions and on the fraction of ions on the outermost shell[4]. In the systems with larger than 10^4 ions, the melting process has not been investigated.

In this paper, we investigate the melting temperature of clusters with 1,000 to 20,288 ions to analyze the mechanism of melting.

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2. COMPUTATIONAL METHOD

We consider the system of particles interacting via the Coulomb potential,

$$v(r) = \frac{q^2}{r}, \tag{1}$$

where q is the charge on a particle and r is the mutual distance. We assume that the external potential in the *Penning-Malmberg* trap is expressed by

$$v_{ext}(r) = \frac{1}{2} k r^2, \tag{2}$$

where k is the constant characterizing a strength of confinement. In this system, the natural scale of length is the unit of Wigner-Seitz radius a_{ws} defined by

$$a_{ws} = \left(\frac{q^2}{k} \right)^{\frac{1}{3}}. \tag{3}$$

This system is characterized by the parameter Γ given by

$$\Gamma = \frac{q^2}{a_{ws} k_B T}. \tag{4}$$

This parameter is a measure of the ratio of the nearest-neighbor Coulomb energy to the thermal energy of particles and signifies the strength of plasma coupling.

We perform molecular dynamics simulations employing the $O(N)$ fast multipole method (FMM)[5]. We recursively divide the system into small cells and compute the contributions from well-separated cells by the multipole and Taylor expansions, applying direct computation only to those from nearby cells. The temperature is controlled by the Nosé-Hoover thermostats[6]. In order to keep the homogeneity of temperature, we attach multiple thermostats each controlling about 500 particles at the same temperature. We anneal the system at the temperature above the melting point of one component plasma (with typical values of around 100) and cool the system slowly enough, reducing the temperature stepwise typically by 15%. The MD simulation had to be run for a sufficiently long time, around 300 plasma periods T_p, to establish a new equilibrium at each temperature:

$$T_p = \frac{2\pi}{\sqrt{3k/m}}. \tag{5}$$

3. RESULTS AND DISCUSSIONS

The specific heat of bulk diverges at the melting point, because melting is first order phase transition. We investigate how specific heat of clusters would behave in the vicinity of temperature considered to be the melting point. The specific heat C_V is calculated by

$$C_V = \lim_{\Delta T \to 0} \frac{\Delta Q}{\Delta T}, \tag{6}$$

where Q is the total heat content of the system.

In Fig.1, we show the specific heat of clusters with 1,000, 10,000, 20,000 and 20,288 ions as a function of temperature, compared with the result of Ref[4]. The melting temperature of the clusters are summarized in Table I. The systems of 1,000, 10,000 and 20,000 ions are shell structured clusters and the system of 20,288 ions is bcc-structured cluster. Except for the system of 20,000 ions, the systems have been heated from $\Gamma = 1000$ to $\Gamma = 100$. The system of 20,000 ions has been cooled from $\Gamma = 100$ to $\Gamma = 300$. The peak temperatures of 1,000 and 10,000 ion clusters in our simulation are nearly the same as those

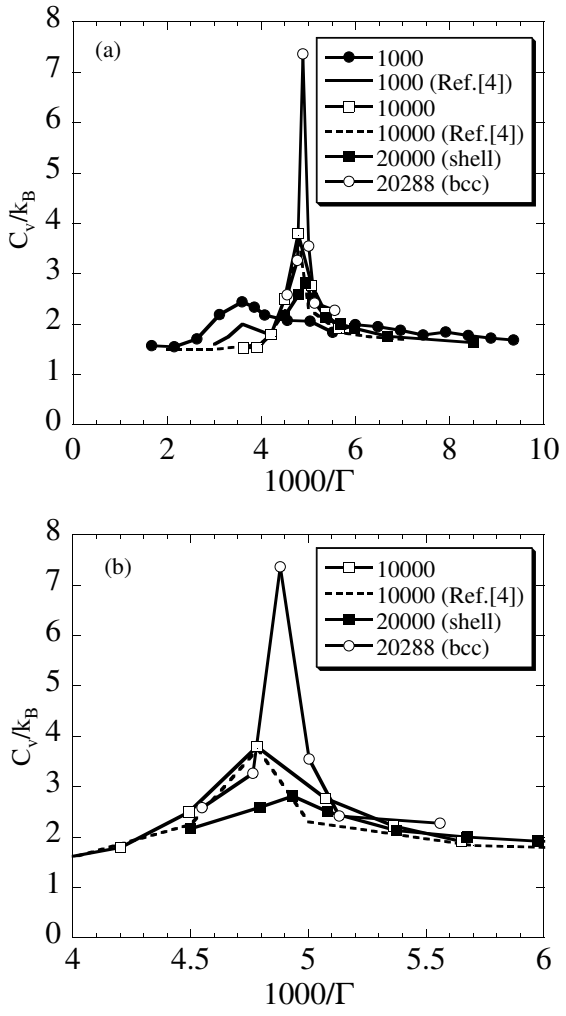

FIG. 1: Specific heat for the system of 1,000, 10,000, 20,000 and 20,288 ions compared with results by Schiffer[4]. Lower figure is the central part of the temperature range of upper one.

TABLE I: The peak temperature of specific heat in the present work compared with Ref.[4].

N	1,000	10,000	20,000 (shell)	20,288 (bcc)
T_{peak} [Γ]	279	209	203	204
$T^{[4]}$	278	209		

in the early work[4]. The anomaly in specific heat gradually weakens with a decreasing number of ions except for the system of 20,000 ions, and the peak is clearly shifting to lower temperatures. For 20,288 ions (bcc structure) and 20,000 ions (shell structure), the peak temperature is in good agreement with each other, but the specific curve of bcc-structured cluster is sharper than that of shell-structured cluster.

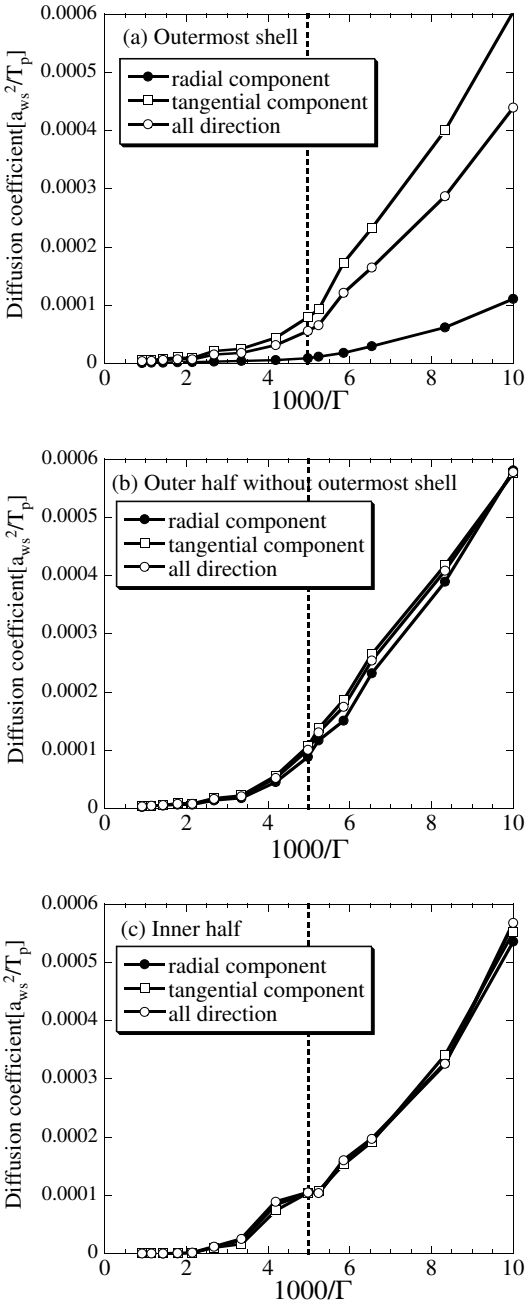

FIG. 2: Diffusion coefficients of (a) outermost shell, (b) the outer half without outermost shell, and (c) the inner half. Open circles are diffusion coefficient, solid circles are that of radial direction, and square are of that tangential direction. The dashed line corresponds to the temperature of the peak of specific heat.

We investigate the diffusion coefficient in order to analyze structural changes. The diffusion coefficient is given by

$$D = \lim_{t\to\infty} \frac{1}{2dt} \left\langle \left|\mathbf{r}\left(t\right) - \mathbf{r}\left(0\right)\right|^2 \right\rangle, \tag{7}$$

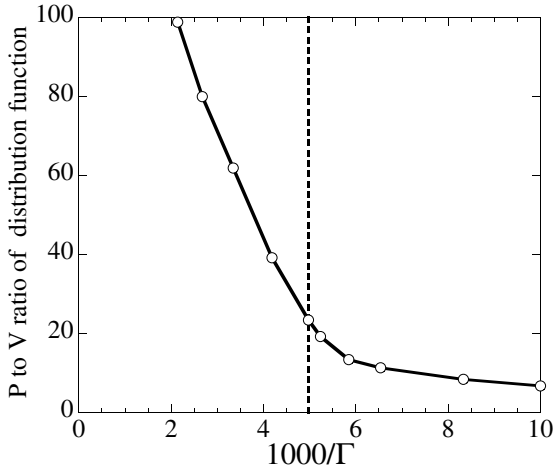

FIG. 3: Peak to valley ratio of pair distribution function. The dashed line corresponds to the temperature of the peak of specific heat.

where d is the dimensionality. In the system of 10,000 ions, ions are divided into three parts, the outermost shell, the outer 50% (in radius) without the surface layer, and inner part. The diffusion coefficient is calculated in each region of ions. In Fig.2, we show the diffusion coefficient which is separated into the tangential component and the radial component. It is found that particles in the outermost shell move easier to the tangential direction than radial direction. This behavior, however, is not observed in other regions. The temperature gradient of tangential component changes near the peak of specific heat in the outermost shell. It is evident that the melting in tangential direction has begun in the outermost shell. The anomaly is also observed in the inner part near the peak temperature, but we have not explained the reason.

We calculate the two-dimensional pair distribution function on the outermost shell, and plot the peak to valley ratio in Fig.3 as a function of temperature. It is also changing near the peak of specific heat. These results suggest the behavior of specific heat is determined by the particles on the outermost shell.

We, finally, investigate rotational movement of shells. We assumed that each shell is rigid, and calculate the rotational kinetic energy K given by

$$K = \frac{1}{2}\frac{L^2}{I}, \tag{8}$$

where L is angular momentum and I is the moment of inertia. Figure 4 shows the rotational kinetic energy of four outer shells and the whole of cluster as a

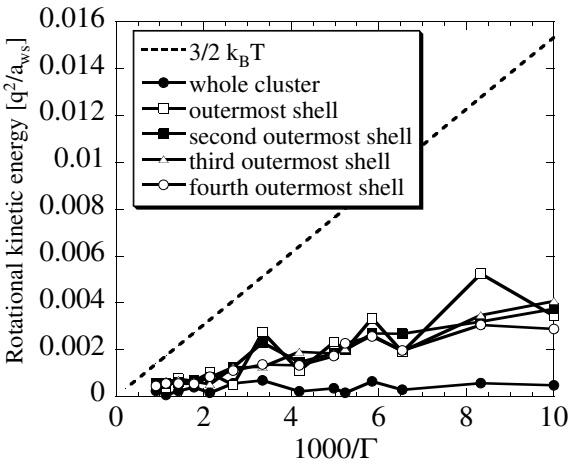

FIG. 4: Rotational kinetic energy of four outermost shells and whole cluster. Dashed line is k_BT of the cluster, and filled circles are the whole cluster.

function of the temperature. The rotational kinetic energy of each shell increases with the temperature, but that of the whole cluster does not change with the temperature. We may conclude the rotational movements of each shell is counterbalanced as a whole. However, there is no anomaly at the temperature of the peak of specific heat. Further investigations seem to be needed.

4. CONCLUSION

We have shown that the melting point of the Coulomb cluster with bcc inner structure is located near that of those clusters not inner shell structure. The specific heat of the former, however, has much sharper peak structure than the latter.

In the shell-structured cluster of 10,000 ions, the tangential diffusion coefficient on the outermost shell shows remarkable increase at the melting point. Diffusions in other direction on this shell or diffusions on inner shells seem to have no anomaly. At finite temperatures, each shell makes the rotational motion with increasing amplitude with temperature.

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