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PHASE TRANSITION OF 2D DIPOLE SYSTEMS IN SCREENING MEDIA

Abstract: The dynamical phase transition of the two-dimensional (2D) electric dipole systems with a background screening charge has been investigated via molecular dynamics simulations. In the simulations three cases of the screening strength, including the bare dipole-dipole interactions, have been examined. In the case where the screening is absent, the quasi-long-range behavior of the bare dipole-dipole interaction potential was handled by implementing the extension of the Wolf method, the gradient shifted force electrostatics. This method enables to efficiently reduce the computational cost of the simulations to $O(N)$with the number of dipoles in the system. The effect of the screening on the course of the solid – liquid phase transitions has been analyzed in detail via the track of the orientational order parameters and polygon order parameter. Results show that the solid – liquid phase transition points of the system occur at lower values of the temperature, or larger values of coupling parameter, due to the screening. We observe that the shift of the melting and freezing points to lower values of the temperature becomes considerable as the strength of the screening is increased.

Key words: two-dimensional (2D) electric dipole systems, screening, the polygon construction method, the Wolf method, the gradient shifted force method.

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2D ДИПОЛЬДЫҚ ЖҮЙЕЛЕРДІҢ ЭКРАНДАЙТЫН ОРТАДА ФАЗАЛЫҚ АУЫСУЫ

Аннотация. Екі өлшемді (2D) электрлік дипольді жүйелердің фондық экрандаушы зарядттар ортасында динамикалық фазалықау ысуы, молекулалық динамика әдісінің көмегімен, зерттелді. Модельдеу кезінде экрандалудың үш жағдайы зерттелді, оның ішін де дипольдардың экрандалусыз өзара әрекеттесуі қарастырылды. Экрандалу болмаған жағдайда, диполь-диполдың өзара әрекеттесу потенциалы, дипольдарға арналған Вольф әдісін қолдану арқылы, өзгертіліп жазылды. Бұл әдіс модельдеудің есептеу шығынын $O(N)$ ретінде тиімді төмендетуге мүмкіндік береді. Экрандалудың кристалл – сұйық фазалық ауысулар процесіне әсері тәртіб параметрлерін есептеу мен көпбұрыштарды құру әдісін қолдану арқылы егжей-тегжейлі зерттелді. Нәтижелер фазалық ауысу нүктелерінің экрандалудың әсерінен төмен температура мандеріне қарай ығысатынын көрсетеді. Сондай-ақ, балқу және кристалдану нүктелерінің төменгі температура мәндеріне ығысуы дипольдардың фондық зарядпен экрандалу күші жоғарылаған сайын артатыны байқалады.

Түйін сөздер: екі-өлшемді (2D) дипольді жүйелер, фондық экрандаушы заряд, көпбұрыштарды құру әдісі, Вольф әдісі.

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ФАЗОВЫЙ ПЕРЕХОД 2D ДИПОЛЬНЫХ СИСТЕМ В ЭКРАНИРУЮЩИХ СРЕДАХ

Аннотация. Динамический фазовый переход двумерных (2D) электрических дипольных систем с фоновым экранирующим зарядом был исследован с помощью молекулярно-динамического моделирования. При моделировании были изучены три случая экранирования, включая диполь-дипольное взаимодействие без экранирования. В случае, когда экранирование отсутствует, квази-дальнодействующий характер диполь-дипольного потенциала взаимодействия был видоизменен путем использования метода Вольфа для диполей. Этот метод позволяет эффективно снизить вычислительные затраты моделирования как $O(N)$с количеством диполей в системе. Влияние экранирования фоновым зарядом диполей на процесс протекания фазовых переходов кристалл – жидкость был подробно проанализирован с помощью расчета параметров ориентационного порядка и использованием метода построения полигонов. Результаты показывают, что из-за экранирования точки фазового перехода кристалл – жидкость дипольных систем смещаются на более низкие значения температур, или на большие значения параметра связи. Также, мы наблюдаем что смещение точек плавления и кристаллизации в сторону более низких значений температур становится более значительным по мере увеличения силы экранирования фоновым зарядом.

Ключевые слова: двумерные дипольные системы, экранирование, метод построения многоугольников, метод Вольфа.

Introduction

This paper presents the investigationofthe phase transitionof two-dimensional (2D) electric dipole systems in the presence of the screening background charge.A 2D many-particle model system with the classical dipole-dipole interaction potential can serve as base for the research and prediction of properties of such systems as 2D complex plasmas [1, 2], polar molecules with low areal density [3, 4], 2D colloidal systems [5-7], and semiconductors [8, 9].

Depending on the degree of screening the dipole-dipole interaction potential changes its behavior from quasi-long range to short range.Therefore, in order to run simulations with bare interaction potential, as well as maintaining the accuracy of calculations, one has to either generate rather large number of dipoles or implement some technique to circumvent the quasi-long-range behavior. Clearly, the first option is not a good choice for ordinary computers, so only option that remains is the modification of the interaction potential.

The quasi-long range character of the pure potential can be circumvented by many methods [10, 12] which can be classified as: implicit (dielectric continuum, static dipolar fields methods) [13, 14], explicit (Ewald method, shifting or truncation of a potential) [15,16], and the combination of the latter two (reaction-field type, fast multipole methods) [17 – 19].The most widely used and preferred methods among the listed are the Ewald method or Ewald based methods, such as the particle-mesh Ewald, particle-particle particle-mesh Ewald, and smooth particle mesh Ewald [15, 20 – 24].However, despite its popularity and common use the Ewald based methods have high computational cost, for given Nparticles at best scaling as $O(N\log N)$[12, 19]. In addition, because of the required artificial periodicity in the Ewald sum it can be problematic in the simulation of some systems [20, 24].

Another method, similar to that of the Ewald method, has been developed by Wolf *et al.* Authors suggested that the effective Coulomb interactions in condensed systems is effectively short ranged and the true interaction potential of a particle with all other particles falls off as $1/r^5$ [25, 26].They also have shown

that this suggestion holds for disordered systems, as well.This method, called the Wolf method, is relatively simple in implementation compared to all other existing methods.It requires only simple modifications to the direct pairwise sum and as a result computational effort scales as $O(N)$, also there is no artificially imposed periodicity as in the Ewald method. In this work, we implement the extension of the Wolf method for higher-order multipoles called gradient shifted force (GFS) electrostatics to simulate dipole systems with the pure interaction potential.

In the next section, we discuss the interaction potential, implementation of the GSF method, and thedescription of simulation method. Then, we present the results and discussions on the phase transition simulations.

Model system

The model system consists of $N = 5041$ point dipoles with equal and parallel dipole moments. Particles areconstrained in square with sidelength $L/a = \sqrt{\pi N}$, where a – the mean interparticle distance, with periodic boundary conditions implemented at the boundaries. The bare pair interaction energy between particles with dipole moment $p$is written as follows

$$U = \frac{p^2}{4\pi\epsilon_0 r^3}. \#(1)$$

As was mentioned earlier Eq. (1) exhibits quasi-long-range behavior, thus we use GFS electrostatics[27] in order to effectively transform it into short-range. The GSF treatment of the pair interaction energy of pure dipoles is given by [27]

$$U_{\text{GSF}} = -\frac{p^2}{4\pi\epsilon_0} u(r), \quad r \le r_c, \#(2)$$

where r_c is the cutoff radius which is set to $r_c = 12\ a$ and $u(r)$ has the form

$$u(r) = \frac{g(r)}{r} - \frac{g(r_c)}{r_c} - (r - r_c)\left(-\frac{g(r_c)}{r_c^2} + \frac{h(r_c)}{r_c}\right).$$

The functions g and h denotes first and second derivatives of expressionerfc$(\alpha r)/r$, where the error function serves as damping and α is the damping parameter which we set $\alpha = 0.2\ a^{-1}$ in our simulations. If we use Smith's functions [28]

$$B_0(r) = \text{erfc}(\alpha r)/r,$$
$$B_1(r) = \frac{1}{r^2}\left(B_0(r) + \frac{2\alpha}{\sqrt{\pi}}\exp(-\alpha^2 r^2)\right),$$
$$B_2(r) = \frac{1}{r^2}\left(3B_1(r) + \frac{4\alpha^3}{\sqrt{\pi}}\exp(-\alpha^2 r^2)\right),$$

then we can write $g$and $h$as

$$g(r) = -rB_1(r),$$
$$h(r) = -B_1(r) + r^2 B_2(r).$$

The total interaction energy is computed as follows [27]

$$U_{\text{tot}} = \frac{1}{2}\sum_{i=1}^{N}\sum_{j=1}^{N} U_{\text{GSF}} + \sum_{i=1}^{N} U_{\text{self}}, \#(3)$$

that is, by including self-term U_{self} in the summation, which is given by

$$U_{\text{self}} = \frac{p^2}{12\pi\epsilon_0}\left(h(r_c) + \frac{2g(r_c)}{r_c} - \frac{2\alpha^3}{\sqrt{\pi}}\right).$$

For values of screening of our interest the screened dipole-dipole interaction potential is short-range, hence there is no need to transform the original form of the potential. In this case, the screened pair interaction energy is given by [29]

$$U = \frac{p^2 \exp(-k_s r)}{4\pi\epsilon_0 r^3}(1 + k_s r), \#(4)$$

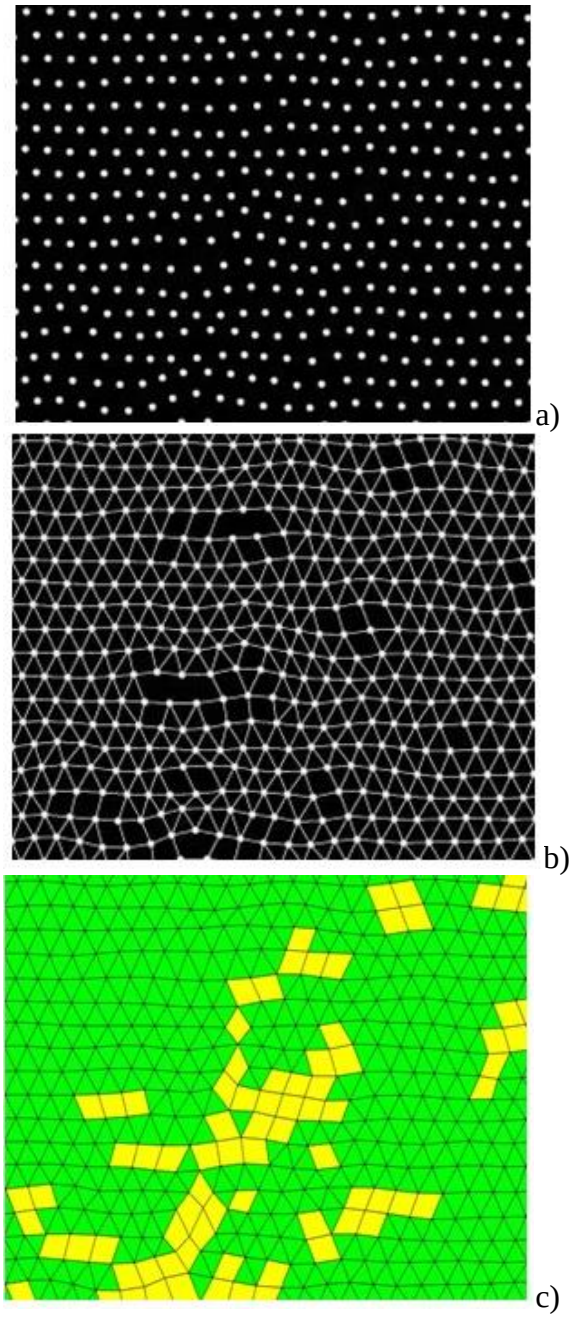
where k_s is the inverse screening length.

The dimensionless coupling parameter $\Gamma_D = p^2/(4\pi\epsilon_0 a^3 k_B T)$ and screening parameter $\kappa = a k_s$ set up the conditions of the model system. In the following, we will focus on three values of the screening parameter $\kappa = 0$, $\kappa = 1$, and$\kappa = 2$.

Results and discussions

We investigate phase transition by means of polygon construction method of Glaser and Clark [30, 31]. This method enables us to track the proliferation of the order and disorder during the phase transition simulations. It does so by means of identifying excess volume in the structure. In order to use the polygon construction method, we first measure the position of each particle creating the particle position map shown Fig. 1 (a). Then, bonds have to be crated between particles, so that long bonds are removed resulting the triangulation map (see Fig. 1 (b)). At the end, depending on the resulting

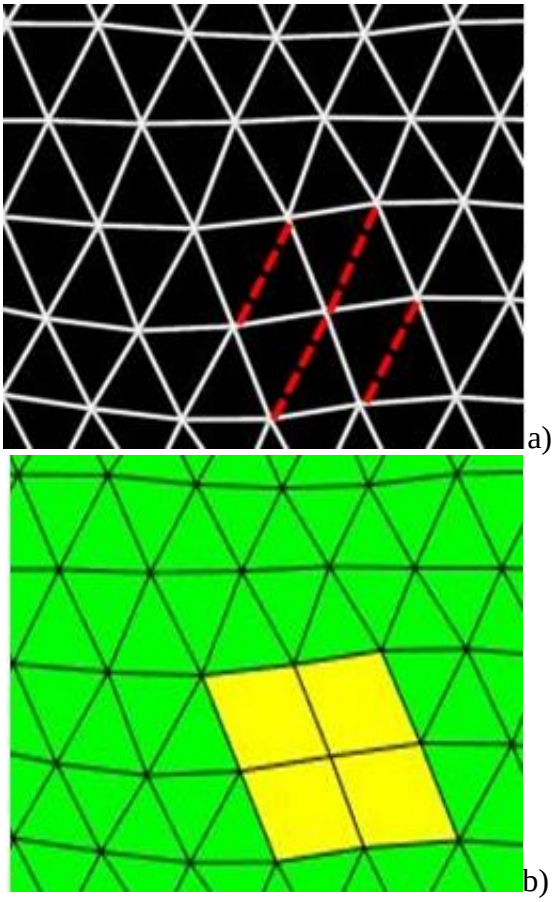
triangulation map the polygon map is created similar to that presented in Fig. 1 (c).



(a) the measurement of particles' positions,
(b) the construction of bonds via the Delaunay triangulation with long bonds removed,
(c) the polygon map that covers entire plane.

Figure 1 – The construction of the polygon map.

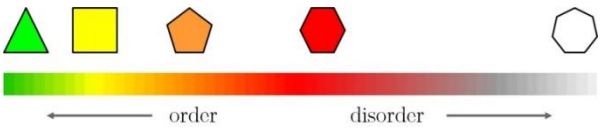
The important criteria of identifying long bonds is done as follows. A bond is considered to be long and removed if the opposite angle to the bond exceeds the value of threshold angle θ_{th} [30, 31]. For example, see Fig. 2 where long bondsare shown as dashed red line. After these bonds are removed the quadrilaterals result identifying defects in the structure. The choice for θ_{th} is arbitrary as long as it varies between 60^{o} and 90^{o}. In our simulations we set $\theta_{th} = 75^{o}$.



By removing bonds that are opposite to large angles from the triangulation map, marked by dashed red lines in (a), we obtain the polygon map in (b)

Figure 2 – The long bond removal.

In the polygon construction method polygons that are triangles termed as geometrical defects. Depending on the degree of disorder the fraction of non-triangular polygons is arranged successively as shown qualitatively in the following diagram.



Two quantities obtained by polygon construction method are the polygon order parameter and vertex fraction. The first one is defined as follows

$$P_p = \frac{N_p}{2N}, \#(5)$$

where N_p is the number of polygons with $p(p = 3,4,5,6)$sides.

Second, the vertex fraction is obtained from knowing the amount of each vertex type, where the vertex type means how the different polygons adjoin around a vertex. Overall, Glaser and Clark have identified twenty-five types of vertices and assigned a letter to each from A to Y [31]. Some of vertex types are presented in the diagram below.

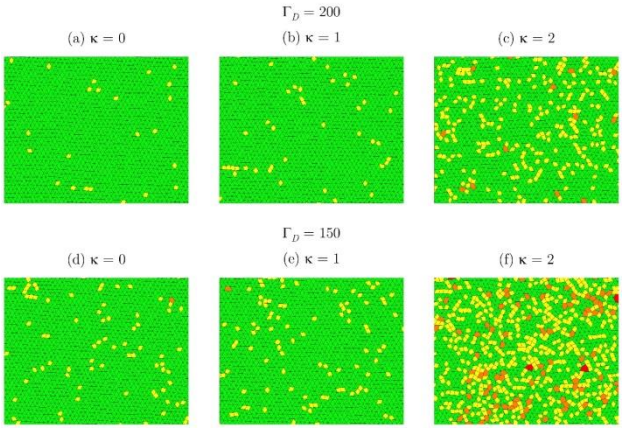
Vertex type	Diagram	Vertex type	Diagram	Vertex type	Diagram	Vertex type	Diagram
A		D		G		J	
B		E		H		K	
C		F		I		L	

The number of each vertex type N_v (where a subscript v denotes the vertex type$A, B, C, \ldots, Y$) is normalized by the total amount of vertices N_v^{tot} to give the fraction of eachvertex in the system structure [32], i.e.

$$V_v = \frac{N_v}{N_v^{\text{tot}}}. \#(6)$$

In the beginning of our simulations we start from the solid state corresponding to $\Gamma_D = 500$. Then, gradually heat the system at rate $dT/dt = 10^{-6}\,\varepsilon\omega_D$, where ω_D – is the characteristic dipole oscillation frequency and $\varepsilon = p^2/(4\pi\epsilon_0 a^3 k_B)$, until Γ_D reaches the value of 50. After that, we run simulations backwards from $\Gamma_D = 50$ to $\Gamma_D = 500$. Simulations are carried out for three values of κ. In the course of the simulation the model system experiences melting and freezing. Our aim is to use polygon construction method to investigate in details the process of phase transition and look for the impacts of screening on melting and freezing.

Fig. 3 illustrates the use of the polygon construction method where vertices match the position of particles and all alike polygons drawn with the same color: triangles – green, quadrilaterals –yellow, pentagons – orange, hexagons – red, and polygons with seven or more sides – white.We are able to observe the emergence of more abundant geometrical defects as a result of stronger screening at specified value of the coupling parameter.Fig. 3 (a) – (c) are sample maps at $\Gamma_D = 200$and Fig. 3 (d) –(f) at $\Gamma_D = 150$.



(a) – (c) are sample maps at $\Gamma_D = 200$ and (d) – (f) at $\Gamma_D = 150$

Figure 3 – Polygon maps for 2D electric dipole systems at two values of the coupling parameter and successive strength of the screening

During the phase transition simulations, we measure polygon order parameters and vertex fractions. The evolution of the latter two with time are presented in Fig. 4 and 5.We can observe that during the heating period the number of triangular polygons start to diminish while other polygons begin to emerge and vice-versa during the cooling period.Triangular polygon order parameter P_3 experiences undergoes decrease in the first stage and increase in the second stage experiencing abrupt jump up and down during the simulation.Quadrilateral polygon order parameter P_4seems like to reach some plateau after sudden jump in its value during the first stage, after the cooling stage starts P_4 stays at plateau before experiencing sudden fall (see Fig. 4 (b)).In the case of pentagon polygon order parameter P_5 we also notice the sudden jump, but here polygons continue to proliferate during the whole heating period, P_5shows the same trend as in first the stage but in reverse direction during the second stage of the simulation (Fig. 4 (c)).Hexagonal polygon order parameter P_6reveals almost steady growth until the cooling period is reached, after that it undergoes steady decrease (Fig. 4 (d)).By observing the impact of screening on polygon order

parameters in Fig. 4 we may conclude that the screening facilitates faster spread of the disorder in the system.

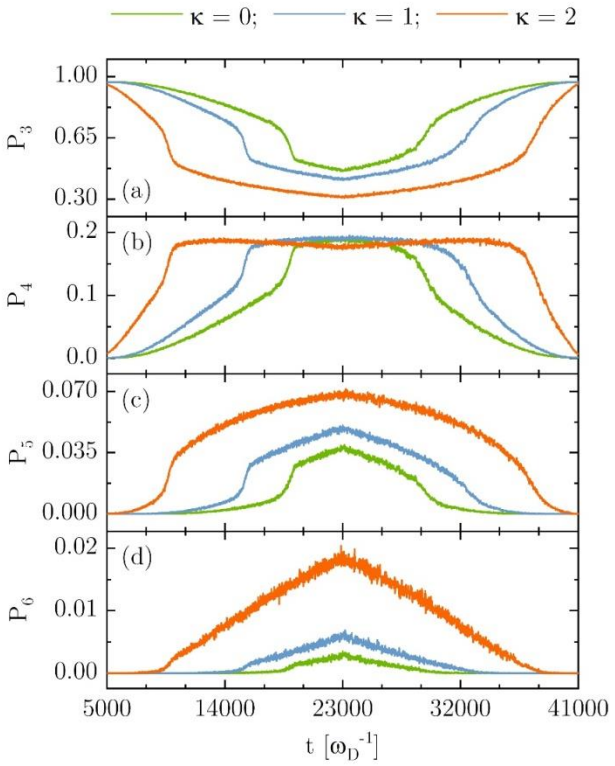


Figure 4 – Evolution of polygon order parameters during heating and cooling periods

Vertex fractions shown in Fig. 5 also confirms above stated fact. It looks like that different vertex types interchangeably grow and diminish during the phase transition simulations. For instance, we see at the beginning of the simulation high fraction of vertex type A, but after heating is progresses V_A decreases while all other vertex types increase. However, at some moment of time vertex types of B, C, E, F reach their maximum value and start to fall indicated byV_B, V_C, V_E, and $V_F$vertex fractions shown in Fig. 5. Also, it is interesting to note that vertex fractions have sudden change in its value at some moment of time. This is clearly observed by V_E and V_F. As will be shown below, that sudden change may be the signal of solid – liquid phase transition.

From the results presented Fig. 5, one can notice the slight non-symmetry between heating and cooling periods. This may suggest that melting and freezing occurs at different values of the system temperature. We can check this by plotting defect fraction (DF) vs. temperature. The DF is defined as follows [33]

$$\mathrm{DF} = \frac{B + C + D + \ldots + X + Y + Z}{N}. \#(7)$$

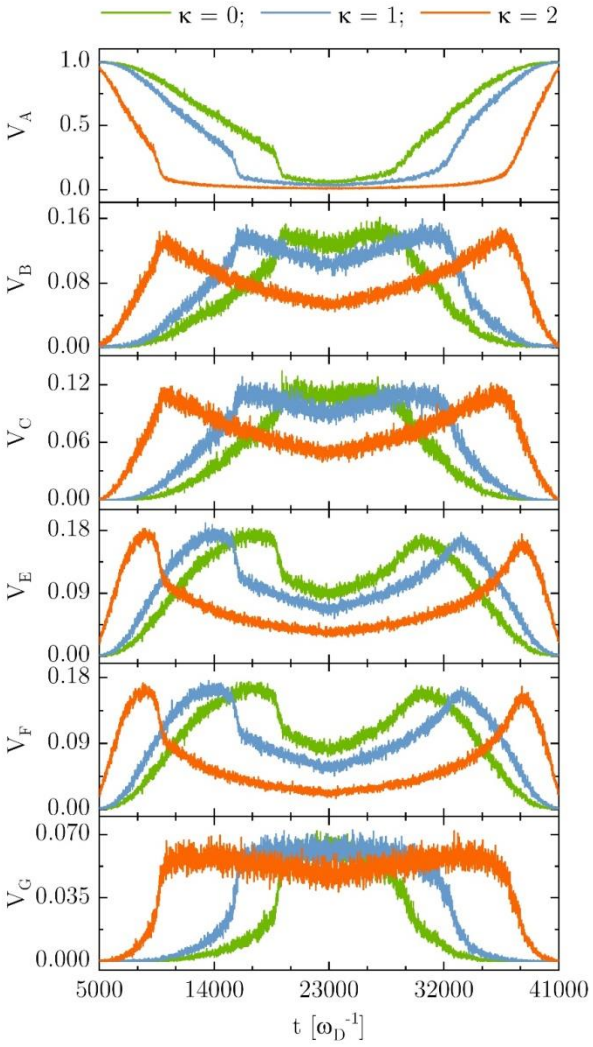


Figure 5 – Evolution of vertex fractions of type A, B, C, E, F, and G during heating and cooling periods

The result is shown in Fig. 6 where arrows indicate direction of simulation. As expected, melting and freezing occurs at different points revealed by the hysteresis. The cause of the hysteresis could be the result of finite temperature change [34]. Thus, in the infinite temperature change no hysteresis should occur.

Apart from hysteresis, we see that screening causes the phase transition point of the system to occur somewhat at larger values of the coupling parameter or lower system temperature. By examining the data in Fig. 6, we conclude that solid – liquid phase transition occurs for $\kappa = 0$ at about $k_B T/\varepsilon = 0.014$, for $\kappa = 1$

at $k_BT/\varepsilon = 0.011$, andfor $\kappa = 2$ at $k_BT/\varepsilon = 0.006$.

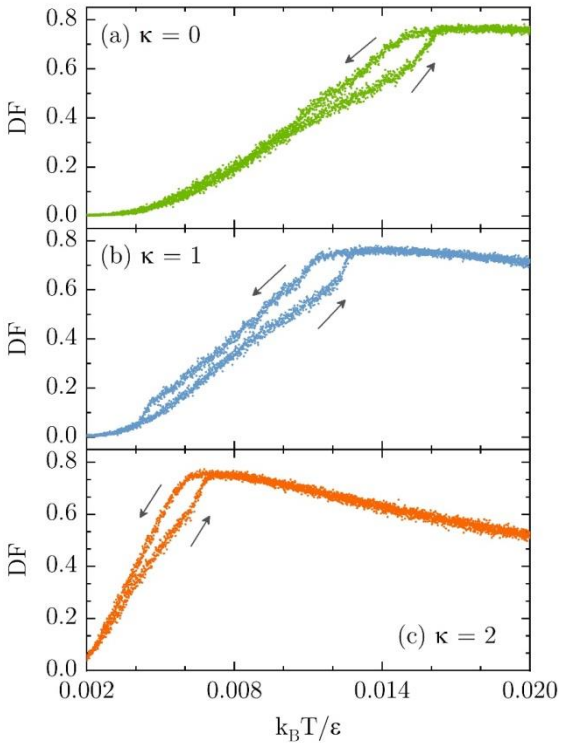


Figure 6 – Defect fraction computed for the 2D system of electric dipoles at three values of screening

One thing which is evident from Fig. 6 is that DF eventually decreases and falls faster with the increasing screening parameter. This drop can be explained by the fact that, as heating progresses more and more non-six type vertices are formed which take greater volume. As a result, much of the space becomes trapped in defect vertices and the total number of vertices decreases giving the plot shown in Fig. 6.

Conclusions

The impact of dipole screening, which due to background charge, on the phase transition points of 2D electric dipoles has been investigated. It has been found via the polygon construction method that screening effectively shifts solid – liquid phase transition points to the lower values of temperature. This in turn implies that the screening facilitates the spread of disorder which has been checked by the phase transition simulations.

It is important to note that bare dipole-dipole interaction potential has been transformed into short-range potential by implementing GFS electrostatics. This method enabled us to perform simulations with less computational effort scaling as $O(N)$.

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