

On the interaction of vacancies in a two-dimensional rare-gas crystal

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Abstract

We study the interaction between vacancies in a two-dimensional van der Waals crystal by molecular dynamics simulation. We find that the vacancy–vacancy interaction is attractive at short distances, but its binding energy is not enough to keep the pair bounded at the melting region where the vacancies are more likely to be created.

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Although an old issue [1,2], the study of point defects in crystals is still an interesting problem [3]. Recently, a series of experiments using video microscopy has succeeded in measuring structural and dynamical properties of defects in colloidal crystals [4], which has provided new experimental tools to study new problems in condensed matter physics [5]. Point defects in solids are of general interest. For instance, for quantum crystals point defects like vacancies and interstitials are supposed to be present in finite concentrations at any nonzero temperature. But, even at zero temperature, they have been speculated to exist as a supersolid [6]. From the theoretical point of view, the role of the point defects to the understanding of the general mechanism of melting, and in particular in a two-dimensional system has been an object of considerable amount of research [7]. Stishov [8] made an extensive analysis of the classical thermodynamics of melting calling attention to a deep investigation on the second-order character of this transition. Toxvaerd [9] studied the liquid–gas interface in a two-dimensional fluid by molecular dynamics. Latter, he calculated the melting line for a two-dimensional Lennard-Jones system using molecular dynamics and showed that melting is a first-order phase transition [10]. Frenkel and McTague [11], using the same method, reached an opposite conclusion which gives support to the theory of

two-stage melting for these two-dimensional systems proposed by Halperin and Nelson [12] and independently by Young [13] (a further elaboration of a theory earlier proposed by Kosterlitz and Thouless [14], the KTHNY theory). Following that, Toxvaerd [15,16] published new molecular dynamic simulations of the same system, showing that it had the same qualitative features of the respective three-dimensional system, a liquid–gas critical point and a first-order phase solid–fluid transition with no evidence of the two-stage melting, and also investigating the size effects on melting and the presence of defects just before the melting. Barker et al. [17] studied the phase diagram of this transition in the same system and again showed the evidence of first-order character and qualitative similarity to the three-dimensional system. Dash [18] noticed that anharmonicity and vacancies can lead to mechanisms that could circumvent the continuous melting. Finally, with the technological advancement, setting up systems with a finite number of particles became a daily reality. Motivated by this, Schweigert et al. [19–22] investigated the role of point defects on melting and crystallization in two-dimensional new systems like confined dust plasma crystals and Coulomb clusters and in quasi-two-dimensional systems like classical bilayer Wigner crystals.

Meanwhile, the experimental investigation of point defects is troubled by the fact that the concentrations of these defects are very small and only near the melting point they achieve their highest values [23,24]. On the other hand, the experimental

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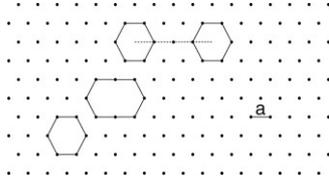


Fig. 1. Two-dimensional triangular lattice with a single vacancy, a divacancy and two vacancies separated by four lattice spacings.

and theoretical study of defect interaction has attracted not too much attention so far [25–28], though its knowledge might provide a better understanding of the microscopic details of the melting mechanism.

This work was motivated by the recent calculations of single point defect interactions in two-dimensional systems like Wigner crystal [27], columnar Wigner crystal [26], and colloidal crystal [28], which show that point defects' interactions in such systems are strongly attractive and must be implied to the investigation of several physical properties [29].

In this work, we apply molecular dynamics simulation to study the energy of formation and interaction of vacancies in a two-dimensional rare-gas crystal. The simulated system is formed by atoms that are treated as “particles” confined to two-dimensions and interacting through a Lennard-Jones central pair-wise potential. The Hamiltonian for such a system is given by

$$H = \sum_i^N \frac{p_i^2}{2m} + \sum_{i<j}^N 4\epsilon \left[\left(\frac{\sigma}{r_{ij}} \right)^{12} - \left(\frac{\sigma}{r_{ij}} \right)^6 \right], \quad (1)$$

where $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$; the first term in the right-hand side is the kinetic energy of the system and the second term is its interaction potential. The energy, length and temperature are in units of ϵ , σ , and ϵ/k_B (k_B is the Boltzmann constant; $\epsilon/k_B = 119.8$ K and $\sigma = 3.4$ Å are the physical parameters of Argon) respectively. The essential feature of this model is that its phase diagram is qualitatively similar in two and three dimensions [30,31]. Therefore, in this two-dimensional simulations the results might be useful for those working on interfaces and solid surfaces, and can qualitatively account for some aspects of the behavior of the three-dimensional crystal, i.e. liquid–gas critical point and first-order character of the transition. Such model is appropriated to study rare-gas crystals since the Lennard-Jones interaction potential describes Argon, Xenon and Krypton well.

In order to model the system with a vacancy, an atom is removed from a site of the most stable two-dimensional lattice which is the triangular lattice. Since it is known that this should cause some relaxation, to avoid any complication we choose to place the vacancy at the center of the simulation box at one of the sites of the triangular lattice. Fig. 1 is a pictorial representation of a single sixfold vacancy, a divacancy and two vacancies separated by four lattice spacing.

The calculation of the energy of formation of a single vacancy was performed by two independent simulations at the same density and temperature: one for the ideal system, i.e. without the defect, and another one for a defective system,

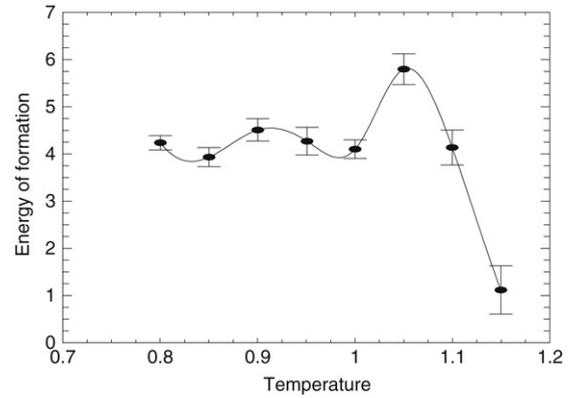


Fig. 2. Energy of formation of a vacancy as a function of temperature at $\rho = 0.9$ in reduced units. Computed for systems with 340 lattice sites.

i.e. with a point defect. For the latter, after placing the defect we rescale the dimension of the simulation box by a factor to reset the system to the original density. By doing this, we prevent the need to correct the calculation of the energy due to changes in the density caused by the inclusion of the defect. The difference between the energies of the defective and the perfect system is the energy of formation of the vacancy. Subsequently, we can define the number of vacancies N_v as the number of atoms minus the number of lattice sites. Therefore, the energy of formation of N_v vacancies in the lattice with N sites is [27]

$$E_v = [e(N + N_v) - e(N)](N + N_v), \quad (2)$$

where $e(n)$ is the energy per atom for a system containing n atoms. For a monovacancy we have $N_v = -1$.

We choose not to enforce any kind of constraint that would restrict the center of each particle to lie within its own Wigner–Seitz cell, as it has been done in the past to allow for local lattice relaxation only [27,32]. It means that the defects are free to move around as well as to change its initial symmetry during the evolution of the system in thermodynamic equilibrium.

We have performed several simulations for lattices with $N = 56, 120, 168, 288, 340, 418, 780$ particles at several densities and temperatures, and a few runs for a larger system to check size effects, with $N = 2340$, which give agreement with the results for the smaller lattice within the statistical error. But, as the system gets larger, the procedure used to calculate the energy of formation of vacancy becomes inadequate since these calculations require high accuracy in order to obtain the differences of energies. Surface effects are eliminated by using periodic boundary conditions. To make the evolution of the classical equations of motion, we have used the fifth-order predictor–corrector algorithm, with a time step varying from 2.5×10^{-2} to 5.0×10^{-3} in reduced units, as it has some scale dependence with the atomic density. The physical quantities are obtained by averaging over 100 000 time steps divided in 10 blocks of 10 000 time steps after the system has reached the equilibrium which was assumed to have occurred during the first 10 000 time steps.

In Fig. 2, we show the energy of formation of a monovacancy as a function of the temperature for a fixed value of the density,

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