PHYSICAL REVIEW B VOLUME 23, NUMBER 9 1 MAY 1981

Melting of a two-dimensional electron lattice

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(Received 9 September 1980; revised manuscript received 17 February 1981)

Melting of a two-dimensional electron lattice is studied using the molecular-dynamics technique. It is found that the system undergoes a first-order phase transition between $\Gamma[=e^2(\pi n)^{1/2}/k_BT]=118-130$. The system exhibits hysteresis in the temperature dependence of total energy and the coefficient of self-diffusion. The change in entropy on melting is found to be $0.3k_B$ per particle.

Recently, there has been a great deal of interest in the phenomenon of melting in two dimensions.[1-4] Grimes and Adams[5] first observed the existence of an electron solid on a liquid-helium surface. Fisher, Halperin, and Platzman[6] have analyzed the experiment and find that the results can be explained on the basis that the electrons form a triangular lattice below the transition temperature. Experimentally, it is found that the liquid-to-solid transition takes place for a value of the coupling constant $\Gamma_m[=e^2(\pi n)^{1/2}/k_BT$, n being the electron density$]=137\pm15$. The classical system of electrons in two dimensions has been studied using Monte Carlo (MC)[7,8] and molecular dynamics (MD) methods[9-11] and the values of Γ_m are in good agreement with the experiment.

In this Communication we report a molecular dynamics study of a classical two-dimensional (2D) electron system. Nature of the liquid-solid transition is examined via structural and dynamical properties. Clear evidence for the existence of hysteresis in the temperature dependence of energy and the constant of self-diffusion lead to the conclusion that the melting of a 2D Coulomb solid is a first-order phase transition. Results are also presented for the specific heat and the velocity autocorrelation function.

MD calculations were performed on a system of 100 electrons on a rigid, uniform, positive background in a rectangular box with periodic boundary conditions. The Hamiltonian of the system is given by

$$H=\tfrac{1}{2}m\sum_i \vec{V}_i^2+\sum_{i>j}\frac{e^2}{|\vec{r}_i-\vec{r}_j|}+U_b\ , \tag{1}$$

where the last term arises from the interaction of electrons with the uniform, positive background. Because of the long-range nature of the Coulomb interaction, we use the method of Ewald summations to take into account the interaction of an electron with an infinite array of periodic images of the other electrons and the uniform, positive background. It should be pointed out here that a rectangle whose sides have the ratio $\sqrt{3}/2$ can accommodate $4M^2$ (where M is an integer) particles on a triangular lattice. Our calculations were performed at a density of 1.477×10^8 electrons/cm^2, in a rectangle of area 0.677×10^{-6} cm^2, and at 20 values of temperatures ranging from 0.19 to 1 K, corresponding to the values of Γ between 180 and 36, respectively. A predictor-corrector method involving up to fifth time derivatives of the positions was employed to integrate the Newton's equations of motion.[12] A time step of 2.5×10^{-12} sec was used in all the calculations, which led to the conservation of energy of 1 part in 50 000 after a 15 000-time-step run. We checked our values of the internal energy and pair correlation functions at $\Gamma=36$ and 90 with those of Gann *et al.* (MC)[8] and Hansen *et al.* (MD),[10] and found them to be in excellent agreement. Our velocity autocorrelation functions at $\Gamma=36$ and 90 are also in excellent agreement with those of Hansen *et al.* In our calculations, at each temperature the system was first thermalized for about 3000 time steps. The results presented in this letter are based on MD runs of 15 000 time steps at each Γ.

In Figs. 1(a), 1(b), and 1(c) we present the results for the temperature dependence of the total energy per particle, coefficient of self-diffusion and the specific heat. The crosses in the figures represent the results for the electron liquid which has been monotonically cooled from a temperature of 1 K ($\Gamma=36$). The solid points are the results for the electron solid which has been monotonically heated starting from a lattice at very low temperatures. The results are independent of the thermal history of the initial lattice configuration. Sizes of crosses and solid points are meant to indicate the degree of uncertainty in the

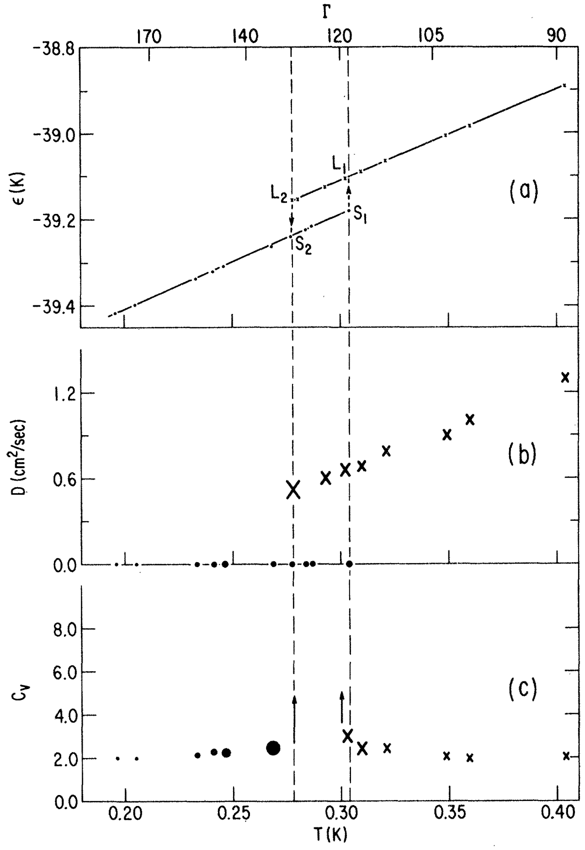

FIG. 1. Temperature dependence of (a) total energy per particle in degrees kelvin; (b) constant of self-diffusion; and (c) specific heat per particle.

values. The region of hysteresis is marked by the two vertical dotted lines. In the temperature range shown in Fig. 1(a) the energy per particle for the solid as well as the liquid is nearly linear with temperature. The point marked S_1 on the lower line (monotonically heated solid) represents the metastable solid at the highest temperature. When this system was heated slightly the system abruptly melted into a liquid marked L_1 via the path indicated by the dotted line. On the liquid line L_2 is the last metastable point which when slightly cooled crystallizes into an electron solid via a path schematically shown by the dotted line. The observed hysteresis in Fig. 1(a) between $\Gamma = 118$ and 130 is clear evidence for the existence of latent heat of melting. Based on our MD calculation we find the value of the latent heat per particle to be 0.09 K and $118 \leq \Gamma_m \leq 130$. From a MC calculation Gann, Chakravarty, and Chester[8] find $\Gamma_m \leq 125 \pm 15$, whereas Morf[11] from an MD calculation gets $\Gamma_m = 130 \pm 10$. While results of these two authors are in good agreement with the experiment as well as our MD calculations, the value of $\Gamma_m = 95 \pm 2$ obtained by Hockney and Brown is somewhat lower.[9]

It should be pointed out here that for a Coulomb system the internal pressure, p, can be obtained in a straightforward fashion from the virial theorem

$$pA = Nk_BT + \frac{1}{2}\left\langle \sum_{i>j} \frac{e^2}{|\vec{r}_i - \vec{r}_j|} \right\rangle . \qquad (2)$$

From this expression and the behavior of energy as a function of temperature, as shown in Fig. 1(a), it is clear that the pressure will also exhibit a discontinuous behavior.

The constant of self-diffusion, D, is calculated for systems on the solid and liquid lines shown in Fig. 1(a). The results for D solid (●) and liquid (×), are shown in Fig. 1(b). Between the hysteresis region marked by the vertical dotted lines, at each temperature D has two values: zero corresponding to the solid phase and about 0.6 cm^2/sec for the liquid phase. It should be emphasized here that the D corresponding to point S_1 is zero whereas when this last metastable state is slightly heated the system melts to L_1 and the constant of self-diffusion jumps from zero to about 0.6 cm^2/sec. In the other instance it is found that the value of D at L_2 is approximately 0.5 cm^2/sec and on cooling the system goes over to S_2 which has a diffusion constant of zero. Such a behavior of the constant of self-diffusion in the hysteresis region is another manifestation of a first-order phase transition.

The specific heat, C_V, of the system is calculated from the fluctuations in kinetic energy in a microcanonical ensemble[13]:

$$C_V = \left[1 - \frac{(\Delta E_k)^2}{N(k_BT)^2} \right]^{-1} . \qquad (3)$$

The results for the solid and liquid phases are shown in Fig. 1(c). It is found that C_V increases when the system approaches the transition region both from the solid and liquid sides. The value of C_V in the hysteresis region shows large variations.

To further demonstrate the nondiffusive and diffusive behavior of the last points on the solid line (S_1) and liquid line (L_2), respectively, we show the behavior of the mean-square displacement $\langle R^2(t) \rangle$ in Fig. 2. It is clear that at S_1 the mean-square displacement does not increase with time and the system is nondiffusive whereas at L_2 the mean-square displacement increases linearly with time and the system shows finite diffusion.

The normalized velocity autocorrelation function

$$Z(t) = \left\langle \sum_i \vec{v}_i(t) \cdot \vec{v}_i(0) \right\rangle \Big/ \left\langle \sum_i \vec{v}_i(0)^2 \right\rangle$$

for the system in the liquid phase at two values of Γ are shown in Fig. 3. The upper curve, $\Gamma = 36$, is for a hot liquid whereas the lower curve, $\Gamma = 130$, is for a system in the hysteresis region. As also observed by Hansen *et al.*,[10] the period of the oscillation in $Z(t)$

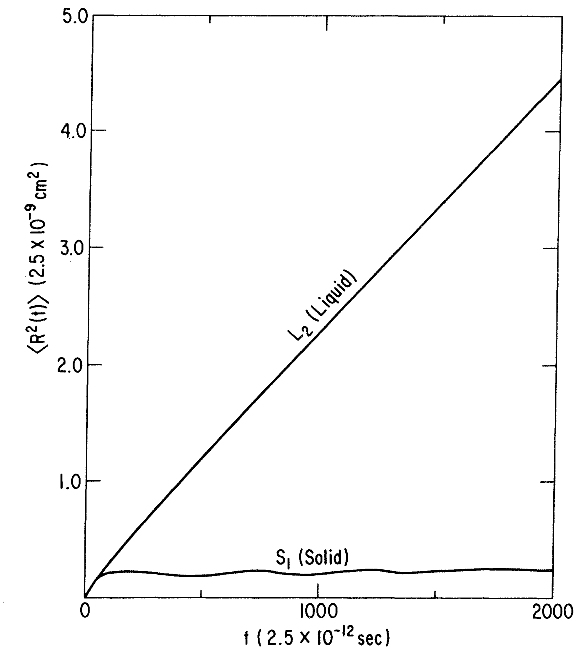

FIG. 2. Mean-square displacement $\langle R^2(t)\rangle = \langle \sum_i [\vec{r}_i(t) - \vec{r}_i(0)]^2/N\rangle$ as a function of time for the last points, L_2 and S_1, on the liquid and solid lines, respectively, in Fig. 1(a).

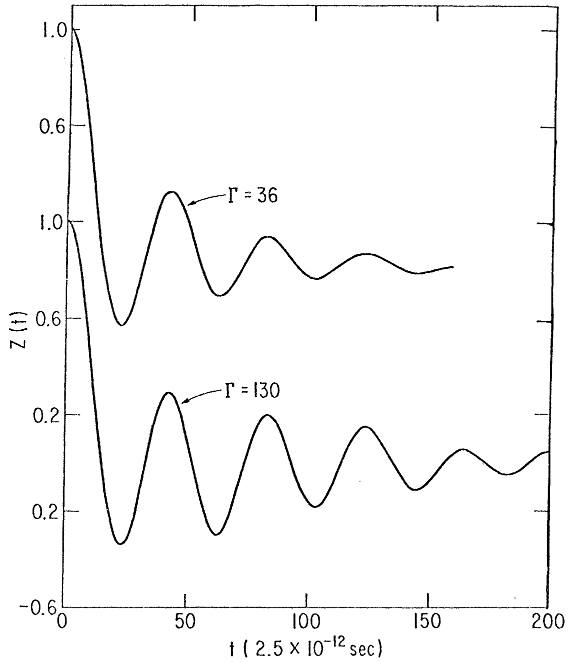

FIG. 3. Time variation of normalized velocity autocorrelation functions at $\Gamma = 36$ (upper curve) and $\Gamma = 130$ (lower curve).

is almost independent of the value of Γ of the system and is not an artifact of the size of the system. Unlike the case of a three-dimensional plasma where oscillation in $Z(t)$ arise from the coupling of the single-particle motion with the well-defined longitudinal-plasmon mode, the situation is somewhat suprising in two dimensions because the characteristic plasmon dispersion goes as the square root of the wave vector.

To check the effect of size of the system on the results described above, calculations were also performed on 256 and 576 particle systems. In all three sizes of the system we find evidence of supercooled liquid and superheated solid. The temperature dependence of total energy for 256 and 576 particle systems is the same as for a 100 particle system, which is shown in Fig. 1. For larger systems too, the latent heat of melting is found to be 0.09 K per particle. The results for the temperature dependence of the constant of self-diffusion and specific heat are again essentially the same irrespective of the size of the system; the results for the larger systems are within the uncertainty indicated in Fig. 1 regarding the results for D and C_V.

In order to further confirm the nature of the phase transition we have studied the homogeneous nucleation in the system. This is done by first lowering the energy of the last supercooled liquid state by an amount that is slightly larger than the latent heat and then the time dependence of instantaneous temperature, $T(t)$, is observed over many thousand time steps. It is found that in the first several thousand time steps $T(t)$ fluctuates around a temperature that is above the lower line for the solid, but then in a small time interval (~ 1000 time steps) the temperature increases and settles lower to a value where the system is exactly on the lower line and it has zero mobility. The observed behavior is unique to systems which undergo first-order phase transition.[14]

Finally, on the basis of the MD results for a classical system of electrons we conclude that the melting of a 2D electron solid is a first-order phase transition. It would indeed be very interesting to experimentally investigate the detailed nature of the melting transition of the electron solid.

We would like to acknowledge many stimulating discussions with Aneesur Rahman, Gianni Jacucci, Alf Sjolander, Kenneth Gray, Sunil Sinha, Lu Sham, Gary Grest, William Kerr, and John Quinn. This work was supported by the U.S. Department of Energy.

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