PHYSICAL REVIEW
LETTERS

VOLUME 43 | 1 OCTOBER 1979 | NUMBER 14

Self-Diffusion in the Two-Dimensional, Classical Electron Gas

J. P. Hansen
Laboratoire de Physique Théorique des Liquides, Université Pierre et Marie Curie, 75230 Paris Cédex 05, France

and

D. Levesque and J. J. Weis
Laboratoire de Physique Théorique et Hautes Energies, Université de Paris XI, 91405 Orsay, France

(Received 6 July 1979)

Molecular-dynamics simulations of the two-dimensional, classical electron gas yield indications for the existence of a self-diffusion constant, in contrast to the case of short-range interactions. The velocity autocorrelation function exhibits marked oscillations in the strong-coupling regime. In order to avoid the Ewald summations of the Coulomb interactions in a periodic system, the electrons are confined on a spherical surface.

The two-dimensional (2D), classical electron gas is a system of charged point particles constrained to move on a surface at a temperature well above the Fermi temperature; charge neutrality is ensured by a uniform background. This system can be considered as a simplified model for electron layers on a liquid helium surface which are being actively studied both experimentally and theoretically.[1] The model ignores, of course, important aspects, like the coupling of the electrons to the capillary waves (ripplons) of the liquid helium surface, but its simplicity renders it more appropriate for the theoretical study of 2D systems. We recall that there are strong theoretical reasons to believe that the self-diffusion coefficient D does not exist for the 2D *hard-disk* fluid,[2-4] due to a slow $1/t$ decay of the velocity autocorrelation function (v.a.f.). The principal motivation of the present work is the investigation of self-diffusion in the 2D electron gas with the help of the "molecular-dynamics" (MD) computer-simulation technique. Static properties of the same model have already been calculated by Monte Carlo (MC) computations,[5,6] while the crystallization of the electron gas has been studied both by MD[7] and MC[6] simulations. The latter computations show that the 2D Wigner transition takes place for a coupling constant $\Gamma = e^2/ak_BT \simeq 10^2$ [where $a = (\pi n)^{-1/2}$ is the radius of the Wigner-Seitz disk and n is the areal density] in agreement with a recent experiment.[8]

We have performed MD simulations for $\Gamma = 36$ and $\Gamma = 90$ which, for a temperature $T = 1$ K, correspond to areal densities $n = 1.5 \times 10^8$ and 9.4×10^8 electrons/cm^2, respectively, typical of experimental situations. Because of the long range of the Coulomb interaction, special care must be taken with the boundary conditions when simulating small systems. The standard treatment, both in three- and two-dimensional simulations, is to use periodic boundary conditions in conjunction with Ewald summations of the interaction of any particle with the infinite array of periodic images of the other particles and the background.[9] We

have explored a different approach by considering a curved, closed surface, avoiding boundaries from the start. The simplest choice is, of course, to place N electrons on the surface of a three-dimensional sphere of radius R.[10] The position of each electron on the sphere is determined by two polar angles or equivalently by a radial unit vector $\vec{u}$. The interaction potential between two electrons, say i and j, is $(e^2/R)\psi_{ij}$, where $\psi_{ij}=\arccos(\vec{u}_i\cdot\vec{u}_j)$, and Newton's equation of motion for the $i$the electron is

$$m\ddot{\vec{u}}_i=-\frac{e^2}{R^2}\sum_{j(\neq i)}\frac{1}{\psi_{ij}^{\,2}\sin\psi_{ij}}[\vec{u}_j-(\cos\psi_{ij})\vec{u}_i]. \quad (1)$$

These equations are equivalent to the equations of motion of N coupled, rigid, three-dimensional rotators[11]; they were solved for systems of $N=104$ and 400 electrons by a finite-difference algorithm, with time steps indicated in Table I. The "natural" temperature-independent unit of time which we use throughout is $\tau=(ma^3/e^2)^{1/2}$; for $n=10^8$ cm^{-2}, $\tau=2.7\times10^{-11}$ sec.

Since it may be argued that the curvature of the spherical surface might introduce systematic differences with respect to the plane periodic system, we have made a detailed comparison of the static properties of our system with the MC results of Gann, Chakravarty, and Chester,[6] who simulated systems of 64 and 100 electrons with periodic boundary conditions. The internal energies are compared in Table I, while the pair distribution functions are compared at $\Gamma=90$ in Fig. 1. The agreement is seen to be excellent, both between results obtained with different system sizes (and hence curvatures), and between the present results and the data of Ref. 6. The structure factor $S(q)$ ($q=ak$) can be calculated from the pair distribution function by a generalized

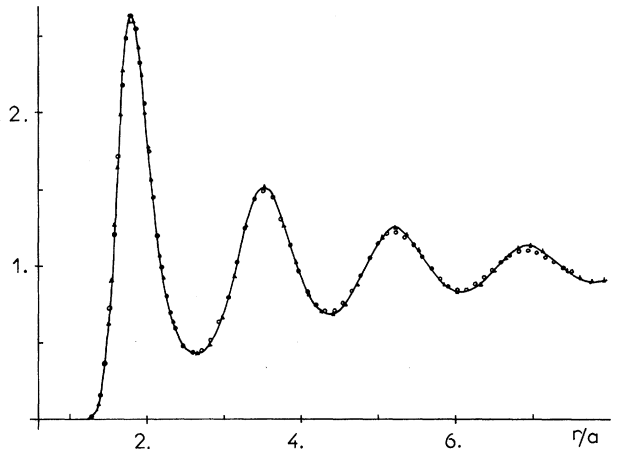

FIG. 1. Pair distribution functions for $\Gamma=90$ for 400 (solid line) and 104 (triangles) particles. The open circles represent the Monte Carlo results of Gann, Chakravarty, and Chester (Ref. 6) using the Ewald method.

Fourier transformation,

$$S(q)=1+2x^2\int_0^\pi[g(x\theta)-1]\sin\theta J_0(x\theta q)\,d\theta, \quad (2)$$

where J_0 denotes the zeroth-order Bessel function and $x=R/a$; this reduces to the ordinary 2D Fourier transform in the limit of zero curvature.

We now turn our attention to the self-diffusion of the electrons. Let $\psi(t)=\arccos[\vec{u}(t)\cdot\vec{u}(0)]$ be the circular arc joining the initial position of an electron to its position at time t on the sphere. We have plotted our results for the function

$$D(t^*)=\frac{x^2\langle\psi^2(t^*)\rangle}{4t^*};\quad t^*=t/\tau, \quad (3)$$

in Fig. 2, for $\Gamma\simeq36$ ($N=104$) and $\Gamma\simeq90$ ($N=104$ and 400). As is clear from the curves, the function $D(t)$ tends, in each case, to a constant after

TABLE I. MD results; N_t is the total number of time steps generated in each run; ω_c is the cyclotron frequency protortional to the externally applied magnetic field B; U denotes the excess internal energy. Note that in a MD computation the coupling constant Γ is not fixed, but is determined by the fluctuating average kinetic energy.

N	Γ	$\Delta t/\tau$	N_t	$\omega_c\tau$	U/Nk_BT	U/Nk_BT (Ref. 5)	D_1	D_2
400	91.2±0.2	0.25	1600	0.0	−99.9±0.2	−99.6	0.014	0.014
104	90.95±0.2	0.25	6400	0.0	−99.2±0.2	−99.3	0.013	0.013
104	90.5±0.2	0.125	6400	0.0	−98.7±0.2	−98.8	0.013	0.014
104	36.1±0.15	0.075	7200	0.0	−38.8±0.15	−38.9	0.047	0.048
104	36.1±0.15	0.075	7200	0.15	−38.8±0.15	−38.9	0.041	0.043
104	35.9±0.15	0.075	7200	3.0	−38.7±0.15	−38.7	0.046	0.046

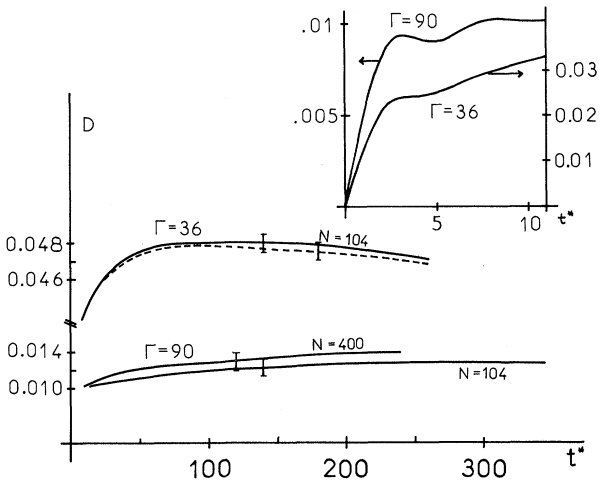

FIG. 2. The function $d(t^*) = x^2\langle\psi^2(t^*)\rangle/4t^*$ for $\Gamma = 36$ and 90. The dashed curve for $\Gamma = 36$ is obtained by applying a magnetic field of $B \simeq 10^5$ G.

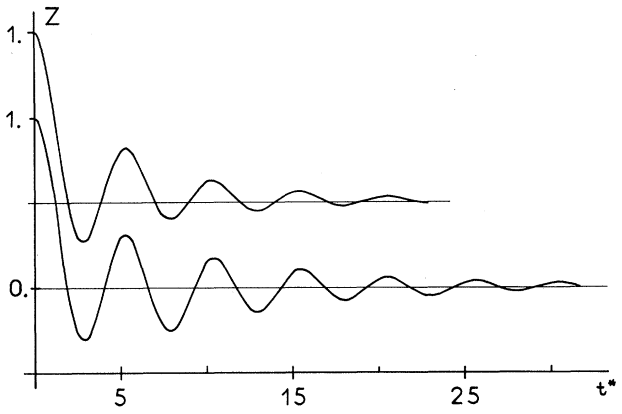

FIG. 3. Normalized velocity autocorrelation function for $\Gamma = 36$ (upper curve) and $\Gamma = 90$ (lower curve).

an initial increase. According to the standard Einstein relation for a two-dimensional system, the constant is just the dimensionless self-diffusion constant D_1:

$$D_1 = \lim_{t^* \to \infty} D(t^*). \tag{4}$$

The asymptotic value D_1 is determined from the horizontal plateau which extends roughly from $t^* = 60$ to $t^* = 200$ for $\Gamma = 36$, and from $t^* = 120$ to $t^* = 300$ for $\Gamma = 90$; the values are listed in Table I. The relative statistical error is estimated to be 10%, and the difference between the results for $N = 104$ and $N = 400$ at $\Gamma \simeq 90$ is well within the combined error bars. Although we cannot rule out that more accurate computations could detect a weak (logarithmic) N dependence of the plateau values D_1, as in the case of hard disks,[4] we conclude that within the present level of accuracy there is no clear indication of such a logarithmic increase and that our data are compatible with the existence of a self-diffusion constant for the two-dimensional electron gas.

There is a decrease of $D(t^*)$ beyond the plateau because D_1 is strictly speaking zero for a closed, finite system, but the finite size is not responsible for the observed saturation of the function $D(t^*)$, since the plateau is reached long before the root-mean-square displacement of an electron approaches half the circumference of the sphere. Converting the reduced (dimensionless) values of the diffusion constant to absolute units, we find at $T = 1$ K the values $D_1 = 0.95$ cm^2/sec at $\Gamma = 90$ and $D_1 = 5.1$ cm^2/sec at $\Gamma = 36$.

In Fig. 3 we have plotted the normalized v.a.f.,

$$Z(t) = \frac{\langle \dot{\vec{u}}(t) \cdot \dot{\vec{u}}(0)\rangle}{\langle |\dot{\vec{u}}(0)|^2\rangle}, \tag{5}$$

obtained for $\Gamma = 36$ and $\Gamma = 90$. The most striking feature is the appearance of well-defined oscillations in $Z(t)$; these oscillations are slightly more damped in the weaker-coupling case, but their period T_Z appears to be essentially the same, in reduced-time units, at $\Gamma = 36$ and 90, i.e., $T_Z \simeq 5.2\tau$ or an angular frequency $\omega_Z \simeq 1.2\tau^{-1}$. The result is qualitatively reminiscent of the three-dimensional classical electron gas (or "one-component plasma"), where $Z(t)$ was found to oscillate at a frequency close to the plasma frequency $\Omega_p = (4\pi e^2 n/m)^{1/2}$.[12] But while the three-dimensional result can be interpreted in terms of a coupling of the single-particle motion to the longitudinal plasma oscillations which are characterized by the well-defined frequency Ω_p in the long-wavelength limit, the two-dimensional result is more surprising since the characteristic $\sqrt{k}$ dispersion of the plasmon mode[1] shows that there is no unique frequency of the 2D longitudinal mode when $k \to 0$. We have checked that the oscillations in $Z(t)$ are not an artifact of the finite size of the simulated system and hence of the curvature, by comparing the v.a.f.'s for $N = 104$ and $N = 400$ at $\Gamma = 90$; the two calculated curves agree very closely.

It is worth pointing out that, because of the curvature of the surface, there is no simple Kubo formula relating the diffusion constant to the time integral of $Z(t)$. In fact, it is readily verified

that

$$\left\langle \sin^2 \frac{\psi(t)}{2} \right\rangle = \langle |\dot{\vec{u}}|^2 \rangle \frac{t}{2} \int_0^t \left(1 - \frac{t'}{t}\right) Z(t')\, dt'. \qquad (6)$$

The standard Kubo formula is recovered from (6) in the zero-curvature limit, when $\sin[\frac{1}{2}\psi(t)]$ can be linearized. However, since $Z(t)$ appears to decay rather rapidly and takes on sizable values only for times such that $\sin^2[\frac{1}{2}\psi(t)] \simeq \frac{1}{4}\psi(t)^2$, it is not surprising that the numerical values for the reduced quantity

$$D_2 = \frac{2}{\Gamma} \int_0^\infty Z(t^*)\, dt^* \qquad (7)$$

lie very close to the diffusion constant, calculated from (3), as can be seen from Table I.

We have completed our MD study of the 2D electron gas by studying the influence of a magnetic field perpendicular to the surface on the self-diffusion of the electrons. The ratio of the Larmor radius over a and the product of the cyclotron frequency $\omega_c = eB/mc$ (where B is in gauss and c is the velocity of light) by the characteristic time τ are both of order one for fields of a few thousand gauss and under typical experimental conditions ($n \simeq 10^8$ cm^{-2}). Hence the cyclotron motion of the individual electrons is characterized by length and time scales similar to those of the interacting electron gas. We have applied magnetic fields such that $\omega_c\tau = 0.15$ (typically $B \simeq 5000$ G) and $\omega_c\tau = 3$ (typically $B \simeq 10^5$ G) at $\Gamma = 36$, and the results are given in Table I. It is seen that the diffusion constant is not affected by the external field, within the statistical uncertainties on D. The v.a.f. is also unchanged.

The main results of the present work are an indication in favor of the existence of a self-diffusion constant in the two-dimensional, classical, electron gas and the appearance of marked oscillations in the v.a.f. at a frequency ω_z which has no obvious explanation. The difference between the self-diffusion in systems with short-range or long-range interactions may be due to the different hydrodynamic behavior of the two classes of fluids; in particular the longitudinal sound mode in the former case is replaced by a plasmon mode with a $\sqrt{k}$ dispersion. Similar differences in the 3D case lead to a very different asymptotic behavior of the v.a.f.[13]

The idea of avoiding periodic boundaries through the use of a spherical surface has been suggested by C. Isenberg at the van der Waals Centenary Conference held in 1973 at the University of Kent at Canterbury, England.

The Laboratoire de Physique Théorique des Liquides is an Equipe associée au Centre National de la Recherche Scientifique; the Laboratoire de Physique Théorique et Hautes Energies is a Laboratoire associé au Centre National de la Recherche Scientifique.

[1]C. C. Grimes, Surf. Sci. 73, 379 (1978).

[2]B. J. Alder and T. E. Wainwright, Phys. Rev. Lett. 18, 988 (1967), and Phys. Rev. A 1, 18 (1970).

[3]M. H. Ernst, E. Hauge, and J. M. J. Van Leeuwen, Phys. Rev. A 4, 2055 (1971); J. R. Dorfman and E. G. D. Cohen, Phys. Rev. A 6. 776 (1972).

[4]W. W. Wood, in *Fundamental Problems in Statistical Mechanics, III*, edited by E. G. D. Cohen (North-Holland, Amsterdam, 1975).

[5]H. Totsuji, Phys. Rev. A 17, 399 (1978).

[6]R. C. Gann, S. Chakravarty, and G. V. Chester, Phys. Rev. B 20, 326 (1979).

[7]R. W. Hockney and T. R. Brown, J. Phys. C 8, 1813 (1975).

[8]C. C. Grimes and G. Adams, Phys. Rev. Lett. 42, 795 (1979).

[9]S. G. Brush, H. L. Sahlin, and E. Teller, J. Chem. Phys. 45, 2102 (1966).

[10]Another possibility would be to use the surface of a torus; the situation would then closely resemble periodic boundary conditions.

[11]J. Barojas, D. Levesque, and B. Quentrec, Phys. Rev. A 7, 1092 (1973).

[12]J. P. Hansen, I. R. McDonald, and E. L. Pollock, Phys. Rev. A 11, 1025 (1975).

[13]H. Gould and G. F. Mazenko, Phys. Rev. A 15, 1274 (1977).