

Novel Electrostatic Attraction from Plasmon Fluctuations

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(Received 19 November 1998)

In this Letter, we show that, at low temperatures, zero-point fluctuations of the plasmon modes of two mutually coupled 2D planar Wigner crystals give rise to a novel long-range attractive force. For the case where the distance d between two planar surfaces is large, this attractive force has an unusual power-law decay, which scales as $d^{-7/2}$, unlike other fluctuation-induced forces. Specifically, we note that its range is longer than the “standard” zero-temperature van der Waals interaction. This result may, in principle, be observed in bilayer electronic systems and provides insight into the nature of correlation effects for highly charged surfaces.

PACS numbers: 61.20.Qg, 05.40.-a, 63.22.+m, 87.16.Dg

Fluctuation-induced forces are ubiquitous in nature (for a recent review, see [1]) and constitute an important contribution to the interactions of many condensed matter systems [2]. The classic example is the Casimir effect [3] in which quantum fluctuations of the electromagnetic field between two parallel conducting plates lead to an attractive force between them. In the context of statistical physics, Fisher and de Gennes [4] have suggested that a similar effect also exists at or near the critical point of a system which is confined between two planes. Other examples include colloid particles immersed in a critical fluid [5], superfluid films [6], liquid crystals [7], and protein inclusions in fluctuating membranes [8]. In general, fluctuation-induced forces arise because external constraints modify the fluctuations of a correlated medium. These interactions, which are usually long ranged, are controlled by thermal fluctuations at finite temperature or quantum fluctuations at low temperature. In this Letter, we present arguments for a long-range attraction, derived from the zero-point fluctuations of the *plasmon* modes associated with two 2D Wigner crystals [9].

Recent attention has focused on fluctuation-induced forces in physical systems that contain charges on surfaces. For example, correlation effects in some 2D electronic systems in semiconductor heterostructures [10]—specifically bilayer systems—could be viewed as a problem of charge fluctuations on surfaces. Charge-fluctuation-induced forces at high temperatures may have another interesting realization in a collection of charged macroions in an aqueous solution of neutralizing counterions, with or without added salt [11,12]. The macroions may be charged membranes, stiff polyelectrolytes such as DNA, or charged spherical colloidal particles. If the surface of macroions is highly charged, most of the counterions in solution condense onto the surfaces [13] and their fluctuations may become important [14].

Specifically, consider a planar surface with charge density en , where e is the electronic charge. A neutralizing

counterion in the solution experiences an (unscreened) electrostatic attractive force of magnitude $4\pi nl_B k_B T$, where $l_B \equiv \frac{e^2}{\epsilon k_B T} \approx 7 \text{ \AA}$ is the Bjerrum length for an aqueous solution of dielectric constant $\epsilon = 80$ (H₂O), k_B is the Boltzmann constant, and T is the temperature. The length scale—the Gouy-Chapman length—at which the thermal energy balances the electrostatic energy, given by $\lambda = 1/(4\pi l_B n)$, defines a layer within which most of the counterions are confined. For sufficiently high charge densities $\lambda \ll L$, where L is the linear size of the macroion, the “condensed” counterions can be considered as a quasi-two-dimensional ideal gas of density n . To capture correlation effects at high temperature, it is sufficient to consider in-plane fluctuations about a uniform charge distribution. Explicit calculations show that charge fluctuations lead to an attractive force between two such plates, which scales as d^{-3} for large separations d [12]. This picture may explain the attractive interaction between two highly charged macroions, observed in experiments [15] and in simulations [16]. Note that this charge-fluctuation-induced force is similar in spirit to the van der Waals interaction, and indeed formally identical to the Casimir force between two partially transmitting mirrors at high temperatures [17]. Although the available experimental results are for high temperature, it is of fundamental interest to understand the low temperature interactions as well. Moreover, although it is unlikely to be relevant for macroions, such considerations may well impact on solid state systems, such as semiconductor bilayers. The purpose of this Letter is to study this charge-fluctuation-induced force at zero temperature.

To this end, we consider a model system composed of two uniformly charged planes, separated by a distance d , each having a charge density of en . Confined to move on them are negative mobile charges, e.g., classical electrons, with particle density n on each of the surfaces, so that the system as a whole is neutral. At sufficiently low temperature, the charges on the surface crystallize into a

2D triangular lattice as in Fig. 1. This occurs when λ is small compared to the average spacing between charges: $\lambda\sqrt{n} \ll 1$, or $\Gamma = e^2\sqrt{\pi n}/\epsilon k_B T$ —the ratio of average Coulomb energy among charges to thermal energy—is sufficiently large [18]. For electrons on the surface of liquid helium, it has been experimentally determined that $\Gamma \sim 100$ [19].

To estimate the attractive force arising from correlation effects at low T , we follow Ref. [20] and consider first the limit of $\Gamma \rightarrow \infty$, i.e., the ground state of the system. In this limit, it is easy to see that, when two classical Wigner crystals experience each other's electric fields, the two lat-

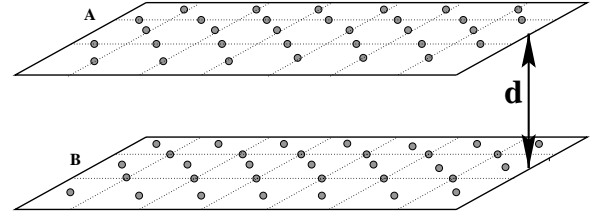


FIG. 1. Two staggered Wigner crystals formed by the condensed counterions.

tices of charges stagger in order to minimize the energy of the system. In this case, the “classical” electrostatic force per unit area of such a charge pattern can be calculated,

$$F_s(d)/A = -\frac{\partial}{\partial d} \left\{ \frac{e^2 n}{\epsilon} \sum_{\mathbf{R}} \frac{1}{\sqrt{|\mathbf{R} + \mathbf{c}|^2 + d^2}} - \frac{(en)^2}{\epsilon} \int \frac{d^2 \mathbf{r}}{\sqrt{r^2 + d^2}} \right\} \approx -\frac{2\pi(en)^2}{\epsilon} e^{-\sqrt{n}d}, \quad (1)$$

for large d , where \mathbf{c} is the relative displacement vector between two lattices of the different plane. Hence, there is a short-range attractive force, which decays exponentially with the lattice constant as the characteristic length scale. In addition to this short-ranged force, we show below that zero-point fluctuations of the plasmon modes (charge fluctuations) also lead to an attractive long-range interaction, analogous to, but scaling differently (in distance) from, the standard zero-temperature Casimir effect. Moreover, at sufficiently low T , this force would still be manifested in the system of classical electrons.

To study the zero-temperature charge-fluctuation-induced force, we first evaluate the phonon spectrum of the system of two coupled Wigner crystals. The positions of the charges are

$$\begin{aligned} \mathbf{r}^A(\mathbf{R}) &= \mathbf{R} + \mathbf{u}^A(\mathbf{R}), \\ \mathbf{r}^B(\mathbf{R}) &= \mathbf{R} + \mathbf{c} + \mathbf{u}^B(\mathbf{R}), \end{aligned} \quad (2)$$

where $\mathbf{u}^{A,B}(\mathbf{R})$ is the small deviation from the equilibrium lattice sites \mathbf{R} . Within the harmonic approximation, the potential may be written as

$$\begin{aligned} \Delta U &= \frac{1}{2} \sum_{i=A,B} \sum_{\mathbf{R}, \mathbf{R}'} K_{\alpha\beta}(\mathbf{R} - \mathbf{R}') u_{\alpha}^i(\mathbf{R}) u_{\beta}^i(\mathbf{R}') \\ &\quad - \sum_{\mathbf{R}, \mathbf{R}'} \Delta_{\alpha\beta}(\mathbf{R} - \mathbf{R}' - \mathbf{c}) u_{\alpha}^A(\mathbf{R}) u_{\beta}^B(\mathbf{R}'), \end{aligned} \quad (3)$$

where repeated indices are summed. Here, $K_{\alpha\beta}(\mathbf{R} - \mathbf{R}') = \delta_{\mathbf{R}, \mathbf{R}'} \sum_{\mathbf{R}''} [\phi_{\alpha\beta}(\mathbf{R} - \mathbf{R}'') + \Delta_{\alpha\beta}(\mathbf{R} - \mathbf{R}'' - \mathbf{c})] - \phi_{\alpha\beta}(\mathbf{R} - \mathbf{R}')$, $\phi_{\alpha\beta}(\mathbf{r}) = \partial_{\alpha} \partial_{\beta} \frac{e^2/\epsilon}{|\mathbf{r}|}$; $\mathbf{r} \neq 0$, and $\Delta_{\alpha\beta}(\mathbf{r}) = \partial_{\alpha} \partial_{\beta} [(e^2/\epsilon)/\sqrt{r^2 + d^2}]$. For a general lattice, the square of the phonon frequencies are the eigenvalues of the dynamical matrix [21], which in this case may be written as

$$\mathbf{D}(\mathbf{k}) = \frac{1}{m} \begin{bmatrix} \mathbf{K}(\mathbf{k}) & -\mathbf{\Delta}(\mathbf{k}) \\ -\mathbf{\Delta}^{\dagger}(\mathbf{k}) & \mathbf{K}(\mathbf{k}) \end{bmatrix}, \quad (4)$$

where m is the mass of the charges and $\mathbf{K}(\mathbf{k})$ and $\mathbf{\Delta}(\mathbf{k})$ are 2×2 matrices whose elements are defined by $K_{\alpha\beta}(\mathbf{k}) = \sum_{\mathbf{R}} K_{\alpha\beta}(\mathbf{R}) e^{-i\mathbf{k} \cdot \mathbf{R}}$ and similarly for $\Delta_{\alpha\beta}(\mathbf{k})$. Using an expansion in reciprocal lattice space, they are explicitly given by

$$K_{\alpha\beta}(\mathbf{k}) = \Delta_{\alpha\beta}(\mathbf{0}) + \frac{2\pi e^2 n}{\epsilon} \left\{ \frac{k_{\alpha} k_{\beta}}{k} + \sum_{\mathbf{G} \neq \mathbf{0}} \left[\frac{(\mathbf{G} + \mathbf{k})_{\alpha} (\mathbf{G} + \mathbf{k})_{\beta}}{|\mathbf{G} + \mathbf{k}|} - \frac{G_{\alpha} G_{\beta}}{G} \right] \right\}, \quad (5)$$

$$\Delta_{\alpha\beta}(\mathbf{k}) = -\frac{2\pi e^2 n}{\epsilon} \left\{ \frac{k_{\alpha} k_{\beta}}{k} e^{-kd} + \sum_{\mathbf{G} \neq \mathbf{0}} \frac{(\mathbf{G} + \mathbf{k})_{\alpha} (\mathbf{G} + \mathbf{k})_{\beta}}{|\mathbf{G} + \mathbf{k}|} e^{-|\mathbf{G} + \mathbf{k}|d} e^{i\mathbf{G} \cdot \mathbf{c}} \right\}, \quad (6)$$

where \mathbf{G} are the reciprocal lattice vectors. In general, to obtain $\omega_j(\mathbf{k})$, the frequency of the j th mode ($j = 1, \dots, 4$), we have to diagonalize $\mathbf{D}(\mathbf{k})$ numerically. This calculation has been performed in Ref. [22]. However, since we are interested in the long-wavelength limit and large distance asymptotics, we approximate $\mathbf{D}(\mathbf{k})$ in the following fashion:

$$\mathbf{D}(\mathbf{k}) \cong \frac{1}{m} \begin{bmatrix} \mathbf{\Delta}(\mathbf{0}) & -\mathbf{\Delta}(\mathbf{0}) \\ -\mathbf{\Delta}^{\dagger}(\mathbf{0}) & \mathbf{\Delta}(\mathbf{0}) \end{bmatrix} + \frac{2\pi e^2 n}{m\epsilon} \begin{bmatrix} \mathbf{D}^0(\mathbf{k}) & -\mathbf{D}^0(\mathbf{k})e^{-kd} \\ -\mathbf{D}^0(\mathbf{k})e^{-kd} & \mathbf{D}^0(\mathbf{k}) \end{bmatrix}, \quad (7)$$

where we have defined the matrix $\mathbf{D}^0(\mathbf{k})$ with elements $D_{\alpha\beta}^0(\mathbf{k}) = \frac{k_{\alpha} k_{\beta}}{k}$. This approximation entails neglecting contributions from higher order terms in k and from nonzero reciprocal lattice vectors, which are exponentially small ($\sim e^{-Gd}$). Therefore, Eq. (7) is a good approximation provided that d is larger than the average spacing between charges on the surface.

Within this approximation, $\mathbf{D}(\mathbf{k})$ can be diagonalized to yield the following dispersion relations:

$$\begin{aligned}\omega_1^2(k) &= 2\Delta, & \omega_2^2(k) &= 2\Delta + \frac{2\pi e^2 n}{m\epsilon} k(1 - e^{-kd}), \\ \omega_3^2(k) &= 0, & \omega_4^2(k) &= \frac{2\pi e^2 n}{m\epsilon} k(1 + e^{-kd}),\end{aligned}\quad (8)$$

where we have chosen $\Delta(\mathbf{0})$ appropriate for staggered triangular lattices: $\Delta_{11} = \Delta_{22} = \Delta$ and $\Delta_{12} = \Delta_{21} = 0$, with $\Delta \sim e^{-Gd}$ [23]. These modes can also be derived by treating the coupling between two isolated Wigner crystals as a perturbation. Mode 1 is one of the two optical modes which correspond to out-of-phase vibrations of the charges in opposite planes. The finite gap at $\mathbf{k} = 0$ vanishes exponentially with d for large distances as also found in Ref. [22]. Mode 3 is the transverse phonon mode, which describes the shear mode of the system, similar to that of a single 2D Wigner crystal. We remark that this approximation gives zero for its frequency, but upon including terms that involve next order in k the dispersion is linear: $\omega_3(k) \sim v_s k$ [22]. Its sound velocity v_s

is roughly a constant—the transverse sound velocity for an isolated Wigner crystal—plus a small correction (exponentially decaying for large d) which arises from the interlayer coupling. In fact, all higher order terms are exponentially damped for large d . Modes 2 and 4 may be interpreted as the out-of-phase and in-phase plasmon modes, respectively. An interesting feature is that the out-of-phase plasmon mode has a gap at $k = 0$ in the presence of the coupling. The in-phase plasmon mode vanishes as \sqrt{k} as $k \rightarrow 0$ and its sound velocity diverges. Physically, the transverse phonon and the in-phase plasmon mode describe the charges in different planes oscillating in phase. We note that their existence has been shown to be a general property for a 2D Coulomb plasma [24] and does not specifically depend on the nature of the underlying Wigner lattice. Thus, the results of this paper are universal, independent of the nature of the ground state.

For the zero-point energy, the dominant modes in the long wavelength limit are the plasmons: modes 2 and 4. Neglecting exponentially small contributions, i.e., $\Delta \rightarrow 0$, we obtain the zero-point energy (relative to the infinite separation) associated with the interactions between the surfaces:

$$\Delta E_0 = \frac{\hbar}{2} \sum_{\mathbf{k}, j} [\omega_j(\mathbf{k}, d) - \omega_j(\mathbf{k}, d \rightarrow \infty)] = A \sqrt{\frac{n\pi e^2 \hbar^2}{2m\epsilon}} \int \frac{d^2 \mathbf{k} \sqrt{k}}{(2\pi)^2} \{\sqrt{1 + e^{-kd}} + \sqrt{1 - e^{-kd}} - 2\}, \quad (9)$$

where A is the area of the planar surface. This leads to an attractive pressure:

$$\Pi_0(d) = -\frac{1}{A} \frac{\partial \Delta E_0(d)}{\partial d} = -\sqrt{\frac{\hbar^2 e^2 n}{m\epsilon}} \frac{\alpha}{d^{7/2}}, \quad (10)$$

where α is a positive numerical constant of order unity, explicitly given by

$$\alpha = \frac{1}{4\sqrt{2\pi}} \int_0^\infty dx x^{5/2} e^{-x} \left\{ \frac{1}{\sqrt{1 - e^{-x}}} - \frac{1}{\sqrt{1 + e^{-x}}} \right\}.$$

Thus, zero-point fluctuations induce a long-range attraction which decays with a novel power law $\sim d^{-7/2}$. This should be contrasted with the usual Casimir-like force $\sim d^{-4}$, which arises from, for example, the acoustic phonon zero-point fluctuations. We note that this power law stems from the two-dimensional nature of charged systems: 2D plasmons do not have a finite gap, as they do in 3D. Note also that all the higher order terms in k , as well as those that we have neglected in our derivation, decay exponentially with d ; therefore Eq. (10) is the dominant term for large distances. For an order of magnitude estimate, assuming $m \sim 10^{-25}$ kg, $n \sim 1/50 \text{ \AA}^{-2}$, $d \sim 10 \text{ \AA}$, and $\epsilon \sim 80$, we find $\Pi \sim 10^{-25} \text{ J/\AA}^3$. This is close to the magnitude of the classical force in Eq. (1): $F_s/A \sim 10^{-24} \text{ J/\AA}^3$, and thus may be just as important under suitable conditions.

To recapitulate, we have argued that there is a long-range force at $T = 0$, derived from the zero-point fluctuations, which must be added to the zero-temperature classical

force. At finite temperatures, an explicit calculation [25] using the Bose-Einstein distribution shows that, at large separations, an additional contribution from the plasmon modes to the attractive force is of the form $\alpha_2 k_B T d^{-3}$, which agrees exactly (even the prefactor α_2) with the high temperatures result of Ref. [12]. Moreover, the effect of finite T on the exponential force is to modify it with a ‘‘Debye-Waller’’ factor, weakening it, and eventually causing it to vanish [25]. Thus, for finite T , we have the following expression for the correlated attractions for a system of two coupled planar Wigner crystals:

$$\Pi(d) = -\alpha_0 e^{-d/a} - \alpha_1 \hbar d^{-7/2} - \alpha_2 k_B T d^{-3}, \quad (11)$$

where $\alpha_{0,1,2}$ are constants and a is the range of the short-ranged attraction, of the order of the lattice constant.

Finally, we comment that experimental observations of our result may prove subtle, as indicated by other examples of phonon-fluctuation-induced interactions. For example, in Ref. [26] the effect of zero-point fluctuations of phonons on the wetting transition of a He thin film was investigated, and the effect was shown to be small. Perhaps, a more appropriate system in which this behavior may be manifested is bilayer quantum well systems. Indeed, recent experimental techniques allow for 2D confinement of electrons, and a 2D plasmon dispersion has been confirmed experimentally in GaAs/AlGaAs heterostructures [27], and may exhibit a fractional power law predicted in this Letter. Although there has been experimental progress in observing single Wigner crystals [28],

the study of interacting Wigner crystals remains an experimental challenge. In any event, the attractive force discussed in this Letter may assist with the conceptual understanding of correlation effects for highly charged surfaces and, more fundamentally, indicates that, for two neutral planes with mobile charges, the zero-point interaction has a universal scaling different from that of the standard Casimir effect.

We would like to thank Professor Walter Kohn, Professor S.J. Allen, and Professor H. A. Fertig for stimulating and helpful discussions. A.L. and P.P. acknowledge support from NSF Grants No. MRL-DMR-9632716, No. DMR-9624091, and No. DMR-9708646, and UC-Biotechnology Research and Education Program. D.L. acknowledges support from Israel Science Foundation Grant No. 211/97. D.L. also would like to thank the MRL at Santa Barbara for its wonderful hospitality.

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