In this Letter, we show that at low temperatures, zero-point fluctuations of the plasmon modes of two mutually coupled 2-D 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.

05.40.-a, 61.20.Qg, 63.22.+m

Fluctuation-induced forces are ubiquitous in nature (for a recent review, see [3]) and constitute an important contribution to the interactions of many condensed matter systems [2]. The classic example is the Casimir effect [2] 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 2-D Wigner crystals [1].

Recent attention has focused on fluctuation-induced forces in physical systems that contain charges on surfaces. For example, correlation effects in some 2-D 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 [3] and their fluctuations may become important [4].

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 $4\pi n l_B k_B T$, where $l_B = \frac{e^2}{\epsilon k_B T} \approx 7\,\text{Å}$ is the Bjerrum length for an aqueous solution of dielectric constant $\epsilon = 80\,(H_2O)$, $k_B$ 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 $d$ [2]. This picture may explain the attractive interaction between two highly charged macroions, observed in experiments [12] and in simulations [13]. 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 [14]. 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 2-D triangular lattice as in Fig. 1. This occurs when \( \lambda \ll 1 \), or \( \Gamma = e^2 \sqrt{\pi n/\varepsilon k_B T} \) — the ratio of average Coulomb energy among charges to thermal energy — is sufficiently large \([8]\). For electrons on surface of liquid helium, it has been experimentally determined that \( \Gamma \sim 100 \) \([13]\).

![FIG. 1. Two staggered Wigner crystals formed by the “condensed” counterions.](image)

To estimate the attractive force arising from correlation effects at low \( T \), we follow Ref. \([20]\) and consider first the limit of \( \Gamma \to \infty \), 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 long-range interaction, analogous to, but scaling differently (in distance) from the standard zero-temperature Casimir effect. Moreover, at sufficiently low \( T \), 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 \( T \), the 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

\[ r^A(\mathbf{R}) = \mathbf{R} + \mathbf{u}^A(\mathbf{R}); \]
\[ r^B(\mathbf{R}) = \mathbf{R} + \mathbf{c} + \mathbf{u}^B(\mathbf{R}), \]

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

\[ \Delta U = \frac{1}{2} \sum_{i=A,B} \sum_{\mathbf{R},\mathbf{R}'} K_{\alpha\beta}(\mathbf{R} - \mathbf{R}') u^A_{\alpha}(\mathbf{R}) u^B_{\beta}(\mathbf{R}') \]
\[ - \sum_{\mathbf{R},\mathbf{R}'} \Delta_{\alpha\beta}(\mathbf{R} - \mathbf{R}' - \mathbf{c}) u^A_{\alpha}(\mathbf{R}) u^B_{\beta}(\mathbf{R}'), \]

where repeated indices are summed. Here, \( K_{\alpha\beta}(\mathbf{R} - \mathbf{R}') = \delta_{\mathbf{R},\mathbf{R}'} \sum_{\mathbf{R}''} \left[ \phi_{\alpha\beta}(\mathbf{R} - \mathbf{R}'') + \Delta_{\alpha\beta}(\mathbf{R} - \mathbf{R}' - \mathbf{c}) - \phi_{\alpha\beta}(\mathbf{R} - \mathbf{R}') \right] \), \( \phi_{\alpha\beta}(\mathbf{r}) = \partial_\alpha \partial_\beta e^{2/\varepsilon |\mathbf{r}|}; \mathbf{r} \neq 0 \), and \( \Delta_{\alpha\beta}(\mathbf{r}) = \partial_\alpha \partial_\beta e^{2/\varepsilon |\mathbf{r}| + d^2} \). 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

\[ \mathbf{D}(\mathbf{k}) = \frac{1}{m} \left[ \begin{array}{cc} \mathbf{K}(\mathbf{k}) & -\Delta(\mathbf{k}) \\ -\Delta^*(\mathbf{k}) & \mathbf{K}(\mathbf{k}) \end{array} \right], \]

where \( m \) is the mass of the charges and \( \mathbf{K}(\mathbf{k}) \) and \( \Delta(\mathbf{k}) \) are \( 2 \times 2 \) 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:

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

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

where \( \mathbf{G} \) are the reciprocal lattice vectors. In general, to obtain \( \omega_j(\mathbf{k}) \), the frequency of the \( j \)th mode \( (j = 1, \ldots, 4) \), 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:

\[ \mathbf{D}(\mathbf{k}) \simeq \frac{1}{m} \left[ \begin{array}{cc} \Delta(0) & -\Delta(0) \\ -\Delta^*(0) & \Delta(0) \end{array} \right] + \frac{2\pi \epsilon^2 n}{m \epsilon} \left[ \begin{array}{cc} \mathbf{D}^0(\mathbf{k}) & -\mathbf{D}^0(\mathbf{k}) e^{-kd} \\ -\mathbf{D}^0(\mathbf{k}) e^{-kd} & \mathbf{D}^0(\mathbf{k}) \end{array} \right], \]

where we have defined the matrix \( \mathbf{D}^0(\mathbf{k}) \) with elements \( D^0_{\alpha\beta}(\mathbf{k}) = \frac{k_\alpha k_\beta}{k} \). 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. \([5]\) 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:

$$\omega_1^2(k) = 2\Delta; \quad \omega_2^2(k) = 2\Delta + \frac{2\pi e^2 n}{me}k \left(1 - e^{-kd}\right);$$
$$\omega_3^2(k) = 0; \quad \omega_4^2(k) = \frac{2\pi e^2 n}{me}k \left(1 + e^{-kd}\right),$$
(8)

where we have chosen $\Delta(0)$ appropriate for staggered triangular lattices: $\Delta_{11} = \Delta_{22} = \Delta$ and $\Delta_{12} = \Delta_{21} = 0$, with $\Delta \sim e^{-Gd}$ [24]. 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: $\omega_3(k) \approx v_s k$ [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 $k \to 0$ 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 confirmed experimentally in GaAs/AlGaAs [25].

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 2 dimensional nature of charged systems: 2-D 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 $m \sim 10^{-25} kg$, $n \sim 1/50 \, \text{Å}^{-2}$, $d \sim 10 \, \text{Å}$, and $\epsilon \sim 80$, we find $\Pi_0 \sim 10^{-25} J/\text{Å}^3$. This is close to the magnitude of the “classical” force in Eq. (1): $F_s/A \sim 10^{-24} J/\text{Å}^3$, 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 [23] 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 $\alpha_2$) 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 [23]. Thus, for finite $T$, we have the following expression for the correlated attractions for a system of two coupled planar Wigner crystals:

$$\Pi(d) = -\alpha_0 e^{-d/a} - \alpha_1 \hbar d^{-7/2} - \alpha_2 k_B T d^{-3},$$
(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. [20] 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 2-D confinement of electrons, and a 2D plasmon dispersion has been confirmed experimentally in GaAs/AlGaAs.
We would like to thank Professors Walter Kohn, S. J. Allen, and H. A. Fertig for stimulating and helpful discussions. AL and PP acknowledge support from NSF grants MRL–DMR–9632716, DMR–9624091, and UC-Biotechnology Research and Education Program. DL acknowledges support from Israel Science Foundation grant 211/97. DL also would like to thank the MRL at Santa Barbara for its wonderful hospitality.

[1] Mehran Kardar and Ramin Golestanian, Rev. Mod. Phys. 71, 1233 (1999); V. M. Mostepanenko and N. N. Trunov, The Casimir Effect and its Applications (Oxford University Press, Oxford, 1997).
[2] J. Mahanty and B. W. Ninham, Dispersion Forces (Academic Press, New York, 1976).
[3] H. B. G. Casimir, Proc. K. Ned. Akad. Wet. 51, 793 (1948).
[4] M. E. Fisher, P.-G. de Gennes, C. R. Acad. Sci. Ser. B 287, 207 (1978).
[5] A. Hanke, F. Schlesener, E. Eisenriegler, and S. Dietrich, Phys. Rev. Lett. 81, 1885 (1998).
[6] H. Li and M. Kardar, Phys. Rev. Lett. 67, 3275 (1991).
[7] A. Ajdari, L. Peliti, and J. Prost, Phys. Rev. Lett. 66, 1481 (1991).
[8] M. Goulian, R. Bruinsma, and P. Pincus, Europhys. Lett. 22, 145 (1993).
[9] E. P. Wigner, Phys. Rev. 46, 1002 (1934).
[10] See, for instance, D. Neilson, S. Swierkowski, J. Szymanowski, and L. Liu, Phys. Rev. Lett. 71, 4035 (1993); L. Zheng and A. M. MacDonald, Phys. Rev. B 49, 5522 (1994); N. P. R. Hill, J. T. Nicholls, E. H. Linfield, M. Pepper, D. A. Ritchie, G. A. C. Jones, Ben Yu-Kuang Hu, Karsten Flensberg, Phys. Rev. Lett. 78, 2204 (1997); S. Das Sarma, E. H. Hwang, Phys. Rev. Lett. 81, 4216 (1998).
[11] O. Spalla and L. Belloni, Phys. Rev. Lett. 74, 2515 (1995); B.-Y. Ha and A. J. Liu, Phys. Rev. Lett. 79, 1289 (1997); R. Podgornik and V. A. Parsegian, Phys. Rev. Lett. 80, 1560 (1998); Ramin Golestanian, Mehran Kardar, and Tammieola B. Liverpool, Phys. Rev. Lett. 82, 4456 (1999).
[12] P. Pincus and S. A. Safran, Europhys. Lett. 42, 103 (1998); Phil Attard, Roland Kjellander, and D. John Mitchell, Chem. Phys. Lett. 139, 219 (1987).
[13] G. S. Manning, J. Chem. Phys. 51, 924 (1969); S. Alexander, P. M. Chaikin, P. Grant, G. J. Morales, P. Pincus, and D. Hone, J. Chem. Phys., 80, 5776 (1984).
[14] F. Oosawa, Polyelectrolytes (Marcel Dekker, New York, 1971).
[15] V. A. Bloomfield, Biopolymers 31, 1471 (1991); R. Podgornik, D. Rau, and V. A. Parsegian, Biophys. J. 66, 962 (1994); A. E. Larsen and D. G. Grier, Nature 385, 230 (1996).
[16] L. Guldbrand, B. Jönsson, H. Wennerström, and P. Linse, J. Chem. Phys. 80, 2221 (1984); S. Marcelja, Biophys. J. 61, 1117 (1992); M. J. Stevens and K. Kremer, J. Chem. Phys. 103, 1669 (1995); N. Grønbech-Jensen, R. J. Mashl, R. F. Bruinsma, and W. M. Gelbart, Phys. Rev. Lett. 78, 2477 (1997); E. Allahyarov, I. D’Amico, and H. Löwen, Phys. Rev. Lett. 81, 1334 (1998).
[17] M. Bordag and K. Schamuhorst, Phys. Rev. Lett. 81, 3815 (1998), and references therein.
[18] Note that for the case of macroions, the formation of Wigner crystal may depend on the valence of the counterions. For monovalent counterions, the assumption of a uniform charge distribution on the surface of the macroion, i.e. the neutralizing background, may not be a good approximation. The reason is that the monovalent counterions stick to the charges on the surface and form dipolar molecules. However, for polyvalent counterions, this does not happen and a Wigner crystal is likely to form. The detailed structure of the ground state as determined by short range effects and valences will be the subject of another study.
[19] C. C. Grimes and G. Adams, Phys. Rev. Lett. 42, 795 (1979); R. C. Gann, S. Chakravarty and G. V. Chester, Phys. Rev. B 20, 326 (1979).
[20] Ioulia Rouzina and Victor A. Bloomfield, J. Phys. Chem. 100, 9977 (1996); B. I. Shklovskii, Phys. Rev. Lett. 82, 3268 (1999); J. J. Arenzon, J. F. Stick, and Y. Levin, Eur. Phys. J. B 7, 79 (1999); V. I. Perel, B. I. Shklovskii, Physica A 274, 446 (1999).
[21] See, for example, N. W. Ashcroft and N. D. Mermin, Solid State Physics (Saunders College Publishing, San Diego, 1976).
[22] G. Goldoni and F. M. Peeters, Phys. Rev. B 53, 4591 (1996); K. Esfarjani and Y. Kawazoe, J. Phys.: Condens. Matter 7 7217 (1995); V. I. Falko, Phys. Rev. B. 49, 7774 (1994).
[23] L. Bonsall and A. A. Maradudin, Phys. Rev. B 15, 159 (1977).
[24] For other lattices, there are slight differences, but the long wavelength behavior remains unchanged.
[25] A.W.C. Lau, Dov Levine, P. Pincus, to be published; see also the first reference in Ref. [24] for a discussion of finite temperature effects on the exponential force.
[26] N. K. Mahale and M. W. Cole, Surf. Sci. 172, 311 (1986).
[27] See, for example, A. Ishihara, Condensed Matter Physics, (Oxford Univ. Press, NY, 1991).
[28] I. N. Harris, H. D. M. Davis, J. F. Ryan, A. J. Turberfield, C. T. Foxon, and J. J. Harris, Europhys. Lett. 29, 333 (1995).