Electrically induced charge-density waves in a two-dimensional electron channel: Beyond the Local Density Approximation

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In a previous paper we predicted that the negative compressibility of a two-dimensional electron gas enables the generation of charge-density waves via the application of a uniform force field (provided no current is allowed to flow) at densities much higher than the critical density for the Wigner crystal transition. Our prediction followed from the minimization of the ground state energy calculated in the local density approximation for both the exchange-correlation energy and the kinetic energy. In this paper we refine our calculation of the energy by including a self-consistent gradient correction to the kinetic energy. Due to the increased energy cost of rapid density variations, we find a much lower critical density for the onset of the charge density wave, but still significantly larger than the expected transition density for the Wigner crystal in the absence of external fields.

I. INTRODUCTION

Spontaneous breaking of translational symmetry in electron liquids is a classic problem in condensed matter physics. The localization of electrons at sufficiently low density was first proposed by Wigner in 1934. [1, 2] He noticed that in this regime the kinetic energy would play a secondary role, allowing the electrons to crystalize in a form that he termed “an inverted alkali metal”. A few decades later Overhauser showed that even at the high densities of (real) alkali metals, exchange and correlation effects would encourage the formation of a charge density wave state (CDW) with wave vector $q \sim 2k_F$, where $k_F$ is the Fermi wave vector.[3] Small ionic displacement would cancel out a large part of the Coulombic energy arising from the non-uniform charge distribution, thus stabilizing the CDW ground state. More recently, broken translational symmetry has been predicted in stripe and bubble phases in electronic systems at high magnetic field, where the magnetic field plays a crucial role in suppressing the kinetic energy of the electrons.[4] Stripes have also been predicted to occur in high-Tc superconductors, of which the copper-oxide superconductors are the most prominent representatives [5].

A different route to stabilizing a CDW state at relatively high density has been recently proposed in Ref. [6]. The idea is to “polarize” a two-dimensional electron channel (2DEC) by applying a uniform in-plane electric field perpendicular to the edges of the channel (see Figure): current flow is prevented by insulating barriers running along the channel. Under “normal” conditions, the electron gas responds to the field by a small rearrangement of its density, resulting in a small accumulation of electrons along one edge and depletion on the other: translational symmetry is not broken on the microscopic scale. The situation may change dramatically when the bulk density $n$ of the 2DEC falls below the value ($r_s \sim 2.1$) for which the thermodynamic compressibility $\kappa = \frac{\partial^2 E}{\partial n^2}$ becomes negative. [7–12] A negative compressibility simply means that, due to the dominance of exchange and correlation effect, the chemical potential of the charge-neutral electron gas decreases with increasing density [13]. By itself, this is not a signal of instability, since the density of the electronic system is stabilized by the requirement of macroscopic charge neutrality. And indeed, in the last few decades, negative electronic compressibility has been experimentally observed in two-dimensional materials through spectroscopy and measurements of quantum capacitance in transition metal dichalcogenide monolayers.[14] carbon nanotubes [15, 16], oxide interfaces, and quantum heterostructures.[7, 16–18] This effect has also been observed in graphene when placed under a strong magnetic field. [12]

Our proposal in Ref. [6] was to combine the negative compressibility with a uniform polarizing electric field. We were surprised to discover that, as soon as the compressibility becomes negative, the equilibrium density in the presence of the field (obtained from a force balance equation which is equivalent to minimizing the ground-state energy in a local density approximation) develops fast oscillations, which break translational symmetry on the microscopic scale. This happens at densities far higher (by orders of magnitude) than the ones usually considered in the context of Wigner crystallization, CDWs etc..

However, there is a problem with this prediction. It is well known that the local density approximation for the energy is justifiable only in the limit in which the density is slowly varying on the microscopic scale, which is the scale of the local Fermi wavelength $k_F^{-1}$. This condition was not satisfied in the calculations of Ref. [6]; on the contrary - the oscillations of the equilibrium density were found to occur on the scale of the screening length $\lambda = \frac{\pi}{k_F} |\frac{\partial n}{\partial e_e}|$, which vanishes at the onset of the instability (meaning that the onset of the instability cannot be determined in a physically reliable manner) and becomes at most as large as $\sim 2.8k_F^{-1}$ at the lowest densities.

The local density approximation is particularly dangerous when applied to the kinetic energy (in which case it is better known as the Thomas-Fermi approximation), whereas its application to the exchange-correlation energy is generally found to work well in ab-initio calculations, for reasons that are quite well understood. In this
Indeed, a significantly smaller value of this value of the Wigner-Seitz radius (regarded as a function of the density $n(r)$, where $r = (x,y)$ is the position in space) has the form

$$\delta \varepsilon_{\text{kin}}[n(r)] = C \frac{\hbar^2}{2m} \frac{\left| \nabla n(r) \right|^2}{n(r)}$$  \hspace{1cm} (1)

where $C$ is a dimensionless constant that can be derived from linear response theory, as detailed in the Appendix. For small density variations the value $C = 1/4$ provides an upper bound to the energy, meaning that by adopting this value of $C$ we almost certainly overestimate the kinetic energy penalty for forming a charge density wave. Indeed, a significantly smaller value of $C$ will be argued below to be physically more accurate.

After including the gradient correction term we repeat the minimization of the total energy of the electron gas in the presence of the force field and find that microscopic translational symmetry is broken at values of the Wigner-Seitz radius $r_0$ that are much larger than 2.1 (the onset of negative compressibility), but still considerably lower than the typical values associated with the onset of Wigner crystallization in the absence of external fields.

$$E[\bar{n}(\bar{x})] = \frac{1}{2} \int_{-1}^{1} d\bar{x} \left\{ \bar{v}(\bar{n}(\bar{x})) + 4C \frac{\bar{L}^2}{\bar{n}(\bar{x})} \right\} + \frac{\bar{V}}{2} \bar{n}(\bar{x}) - \bar{L} \int_{-1}^{1} d\bar{x} \delta \bar{n}(\bar{x}) \delta \bar{n}(\bar{x}') \ln |\bar{x} - \bar{x}'| \right\},$$  \hspace{1cm} (3)

where $\bar{E}[\bar{n}(\bar{x})]$ is the functional derivative of the energy with respect to the density deviation $\delta \bar{n}(\bar{x})$, subject to the constraint (2). This gives us the equation

$$\frac{\delta E[\bar{n}(\bar{x})]}{\delta \bar{n}(\bar{x})} = \mu,$$  \hspace{1cm} (4)

where $\frac{\delta E[\bar{n}(\bar{x})]}{\delta \bar{n}(\bar{x})}$ is the functional derivative of the energy functional with respect to $\bar{n}(\bar{x})$, and $\mu$ is a constant Lagrange multiplier required to satisfy the constraint (2). Alternatively, we can write

$$\frac{d}{d\bar{x}} \frac{\delta E[\bar{n}(\bar{x})]}{\delta \bar{n}(\bar{x})} = 0.$$  \hspace{1cm} (5)

Carrying out the prescribed operations and keeping only terms linear in $\delta \bar{n}$ (under the assumption that $\delta \bar{n} \ll \bar{n}_0$) we arrive at the integrodifferential equation

$$\bar{v}''(\bar{n}_0) \frac{\delta \bar{n}(\bar{x})}{2L} - 4C \frac{\delta \bar{n}(\bar{x})}{\bar{n}_0 \bar{L}^3} - \int_{-1}^{1} dx' \delta \bar{n}(\bar{x}') \frac{\delta \bar{n}(\bar{x}')}{\bar{x} - \bar{x}'} = -\frac{\bar{V}}{4L},$$  \hspace{1cm} (6)

where $\bar{v}''(\bar{n}_0)$ is the second derivative of the energy density evaluated at the background density. This second derivative is related to the compressibility $K(n)$ by the
well-known formula $\epsilon''(n) = 1/(n^2 K(n))$ and becomes negative (passing through zero) for $r_s$ greater than about 2.1. In the above equation $\bar{x}$ is restricted to the interval $|\bar{x}| < 1$ and the integral is evaluated according to the Cauchy principal value prescription (denoted by a strike across the integral sign). For $C = 0$ Eq. (6) reduces to the force balance equation used in Ref. [1]. Notice that the equation is linear in $\delta n$, so the induced density is directly proportional to the force applied, $F = -V/L$.

III. APPROXIMATE SOLUTION

In the limit of large channel width (i.e., $L \to \infty$) it may appear that only the Hartree term on the left side of Eq. (6) is relevant. Neglecting the remaining terms (we have omitted a term proportional to $C/L^3$, which is negligible in the limit $L \to \infty$).

The consistency of our Ansatz requires the coefficient of $\cos(\bar{q}\bar{x})$ to vanish, thus determining $\bar{q}/L$ as the positive solution of the equation

$$\frac{\epsilon''(\bar{n}_0)}{2\pi} \left( \frac{\bar{q}}{L} \right) + \frac{4C}{\pi \bar{n}_0} \left( \frac{\bar{q}}{L} \right)^3 + 1 = 0. \quad (11)$$

would give the elegant classical solution

$$\delta n(\bar{x}) = -\frac{V}{4L \pi \sqrt{1 - \bar{x}^2}}. \quad (7)$$

This conclusion is correct only as long as the density remains slowly varying on the macroscopic scale ($L$). In our previous paper we showed that a negative compressibility can induce rapid variations of the density on a microscopic scale ($a$), so that the derivatives of the density on the left hand side of Eq. (6) must be taken into account. Unfortunately, rapid variation of the density also means that the validity of the local density approximation for the energy (i.e., the first term on the the left hand side of Eq. (6)) is dubious. Hence the importance of including the gradient correction term, which strongly discourages such rapid variations by imposing a large energy penalty.

Based on these considerations, we seek a solution of Eq. (6) in the form

$$\delta n(\bar{x}) = -\frac{V}{4L} \left\{ \frac{x}{\pi \sqrt{1 - x^2}} + f \sin (\bar{q}x) \right\}, \quad (8)$$

that is to say the classical solution plus an oscillating term with dimensionless wave vector $\bar{q} = qL/2 = (qa)L/2$. We assume $q > 0$, while the real amplitude $f$ can have either sign as needed. This Ansatz has been shown in Ref. [6] to yield excellent results vis-a-vis the exact numerical solution of the integro-differential equation.

We substitute Eq. (8) into Eq. (6) and focus on the region $|\bar{x}| \ll 1$ (which, we emphasize, can be macroscopically large if $L$ is large). In this region we have

$$\int_{-1}^{+1} dx' \frac{\sin(\bar{q}x')}{x - x'} \simeq -\pi \cos(\bar{q}x) + \pi - 2Si(\bar{q}), \quad |\bar{x}| \ll 1, \quad (9)$$

where $Si(\bar{q}) \equiv \int_0^{\bar{q}} \frac{\sin(t)}{t} dt$ is the Sine Integral function and the strike across the integral sign mandates that we take the principal value of the integral. Thus, we arrive at the following equation for $\bar{q}$ and $f$:

$$\left\{ \frac{\epsilon''(\bar{n}_0)}{2\pi} \left( \frac{\bar{q}}{L} \right) + \frac{4C}{\pi \bar{n}_0} \left( \frac{\bar{q}}{L} \right)^3 + 1 \right\} \cos(\bar{q}\bar{x}) - \left[ 1 - 2Si(\bar{q})/\pi \right] f + \frac{\epsilon''(\bar{n}_0)}{2\pi^2 L} = 0 \quad (10)$$

Notice that $\bar{q}/L = qa/2$ so we expect a solution with $qa/2$ on the order 1, independent of $L$ in the limit $L \to \infty$. This corresponds to a microscopic charge density wave.

Given that $C > 0$, it is immediately evident that a solution can exist only if the compressibility is negative, i.e. for $\epsilon''(\bar{n}_0) < 0$. The plain local density approximation $C = 0$ yields a solution for any density such that

![Figure 1. Critical Wigner-Seitz radius $r_{sc}$ (blue curve) and the wave vector of the CDW instability at the critical Wigner-Seitz radius $q_c/(2k_Fc)$ (red curve) vs coefficient of the gradient correction term $C$.](image-url)
\[ \tilde{\epsilon}''(\bar{n}_0) < 0, \] but then the wave vector of the solution diverges when the density is close to the critical value for which \( \tilde{\epsilon}'' \) vanishes. On the other hand, a sufficiently large value of \( C \) may completely prevent the existence of a solution, by making the left hand side of Eq. (11) always positive for \( \bar{q} > 0 \).

For a given value of \( C \) a solution of Eq. (11) first appears at a critical Wigner-Seitz ratio \( r_{sc} \) and wave vector \( \bar{q}_c \) such that

\[ \frac{\tilde{\epsilon}''(r_{sc})}{2\pi} + 12C r_{sc}^2 \left( \frac{\bar{q}_c}{L} \right)^2 = 0, \] (12)

i.e., when the zero of the left hand side of Eq. (11), regarded as a function of \( \bar{q} \), yields the minimum of that function. Combining Eqs. (11) and (12) we find

\[ \frac{\bar{q}_c}{L} = -\frac{3\pi}{\tilde{\epsilon}''(r_{sc})}, \quad C = \left( \frac{\tilde{\epsilon}''(r_{sc})}{6\pi^2 r_{sc}^3} \right)^3. \] (13)

It is convenient to express the wave vector \( q_c \) in terms of the Fermi wave vector \( k_{Fc} \) at the critical density. Noting that \( k_{Fc}a = \sqrt{2}/r_{sc} \) we get

\[ \frac{q_c}{k_{Fc}} = -\frac{6\pi r_{sc}}{\sqrt{2}\tilde{\epsilon}''(r_{sc})}. \] (14)

In Fig. 1 we plot \( q_c/k_{Fc} \) and the critical Wigner-Seitz radius \( r_{sc} \) as functions of \( C \). Clearly the plot is meaningful only for \( \tilde{\epsilon}''(r_{sc}) < 0 \), i.e., for \( r_{sc} > 2 \). Numerical values of \( \tilde{\epsilon}''(r_{sc}) \) are taken from Ref. [19]. We note in passing that an excellent approximation (for the present purpose) to \( \tilde{\epsilon}''(r_{sc}) \) for \( r_{sc} > 2 \) is

\[ \tilde{\epsilon}''(r_{sc}) \simeq -1.35\pi(r_{sc} - 2), \quad r_{sc} > 2. \] (15)

For a given value of \( C \) in the range \( 0 < C < 1/4 \), where \( C = 0 \) neglects the gradient correction and \( C = 1/4 \) is an upper bound (see next section) the blue curve in Fig. 1 allows us to read the value of the critical \( r_{sc} \), which is then used in Eq. (14) to determine \( q_c/(2k_{Fc}) \) (red curve). We see that even in the most unfavorable case, \( C = 1/4 \), our model predicts a charge density wave instability with \( q_c \approx 3.38k_{Fc} \) and critical \( r_{sc} \approx 27 \), which is smaller than the critical \( r_{sc} \) for Wigner crystallization in 2D. However, there are good reasons to believe that the appropriate value of \( C \) is significantly smaller than 1/4. Indeed, a self-consistent gradient correction, described in the next section, produces a lower value of \( r_{sc} \). For completeness, we also calculate the amplitude \( f \) of the charge density wave (normalized by \( \tilde{V}/(4L) \)). This is given by

\[ f = \frac{\tilde{\epsilon}''(\bar{n}_0)}{2\pi^2 L[1 - 2Si(\bar{q})/\pi]} \] (16)

In the limit of large \( L \) we make use of the limiting form

\[ [1 - 2Si(\bar{q})/\pi] \rightarrow \frac{2}{\pi\bar{q}} \cos \bar{q}, \quad \bar{q} \gg 1, \] (17)

\[ \text{Figure 2. Plot of } \frac{-m}{\pi^2}\left[\chi_0^{-1}(q) - \chi_0^{-1}(0)\right] \text{ (blue-solid line) and its large-} q \text{ asymptote } \frac{q^2}{2k_F^2} \text{ (red-dashed line) vs } q/k_F. \]

\[ \text{Inset shows } C(q) \text{ from Eq. (22) vs } q/k_F. \]

\[ \text{to arrive at} \]

\[ f = \frac{\tilde{\epsilon}''(\bar{n}_0)(qa)}{8\pi \cos(qL/2)} \] (18)

which is finite, but exhibits a wild non-analytic dependence on \( L \).

\[ \text{IV. SELF-CONSISTENT GRADIENT CORRECTION} \]

In this section we analyze more closely the nature of the gradient correction to the kinetic energy density of a weakly inhomogeneous 2DEG \( (\delta \bar{n}(\mathbf{r}) \ll \bar{n}_0) \). To second order in \( \delta \bar{n} \), the kinetic energy density relative to the homogeneous state is given by

\[ \epsilon_{\text{kin}}[\delta \bar{n}] = \frac{1}{2A} \int \frac{d^2q}{(2\pi)^2} \chi_0^{-1}(q)|\delta \bar{n}(\mathbf{q})|^2, \] (19)

where \( \chi_0(q) \) is the static Lindhard function (i.e., the static density-density response function of the noninteracting 2DEG), \( \delta \bar{n}(\mathbf{q}) \) is the Fourier transform of \( \delta \bar{n}(\mathbf{r}) \) at wave vector \( \mathbf{q} \), and \( A \) is the area of the system. The static Lindhard function is known analytically:

\[ \chi_0(q) = -\frac{m}{\pi\hbar^2} \left[ 1 - \Re \sqrt{1 - \frac{4k_F^2}{q^2}} \right]. \] (20)

It is well-known that the local density approximation for the kinetic energy is obtained (to second in \( \delta \bar{n} \)) by ignoring the \( q \)-dependence of \( \chi_0(q) \), i.e., by setting \( \chi_0^{-1}(q) = \chi_0^{-1}(0) \) in Eq. (19). The exact correction to
the local density approximation for the kinetic energy is therefore given by
\[
\delta \epsilon_{\text{kin}}[\delta n] = -\frac{1}{2A} \int \frac{d^2 q}{(2\pi)^2} \left[ \chi_0^{-1}(q) - \chi_0^{-1}(0) \right] |\delta n(q)|^2 ,
\]
(21)
The quantity \(-[\chi_0^{-1}(q) - \chi_0^{-1}(0)]\) is plotted in Fig. 2 (in units of \(m/(\pi\hbar^2)\)). It is strictly zero for \(q < 2k_F\) and remains always lower than the parabola \(q^2/2k_F^2\), to which it tends for \(q \to \infty\). It is easy to verify that the gradient correction of Eq. (1), with \(C = 1/4\) is obtained precisely by approximating \(-[\chi_0^{-1}(q) - \chi_0^{-1}(0)]\) by its high-q limit \(\pi k_F^2 q^2/2mk_F^2\). Therefore Eq. (1), with \(C = 1/4\), definitely overestimates the kinetic energy cost of the inhomogeneity. From Fig. 2 we also see that density waves with wave vectors smaller than \(2k_F\) are adequately described by the local density approximation. Therefore the choice \(C = 1/4\) is inconsistent, since it predicts a density wave with \(q \simeq 3.38k_F\) (at \(r_s \simeq 27\)), which in turn would yield a value of \(C\) smaller than 0.2 as can be seen in the inset of Fig. 2. However, the choice \(C = 0\) would also be inconsistent, since it would predict wave vectors much larger than \(2k_F\), which would imply \(C = 1/4\). These considerations lead us to propose that the value of the coefficient \(C\) in the kinetic gradient correction can be chosen consistently with the value of \(q_c\) that it predicts. In other words, we set
\[
C(q) = \frac{m}{\pi \hbar^2} \frac{\chi_0^{-1}(q) - \chi_0^{-1}(0)}{2(q/k_F)^2} ,
\]
(22)
such that \(C \to 1/4\) for \(q \to \infty\) and \(C = 0\) for \(q < 2k_F\).

Substituting \(q_c\) from Eq. (14) in the above expression, and setting it self-consistently equal to \(C\) given by Eq. (13) yields the self-consistent value of the critical \(r_s\) as \(r_s \simeq 22.3\), with \(q_c \simeq 3.49k_F\) and \(C_c = 0.186\).

V. DISCUSSION AND OUTLOOK

The calculations presented in this paper strengthen the conclusions of Ref. [6]. The prediction that a uniform electric field would induce microscopic translational symmetry breaking at density for which the 2DEG compressibility is negative is confirmed. In addition, we are able to pinpoint a critical value of \(r_s\) and the wave vector of the induced charge density wave. We emphasize that the predicted critical value of \(r_s\) is lower than the one commonly associated with Wigner crystallization and should therefore be more easily accessible in experiments. Our gradient approximation to the kinetic energy density overestimates the energy penalty associated with a density modulation: it is therefore expected that a more accurate calculation, based on a self-consistent solution of the Kohn-Sham equation with a local density approximation for the exchange-correlation energy would lead to similar conclusions. Going beyond the local density approximation for the exchange-correlation energy would probably introduce small changes of a quantitative character.

While the results for critical \(r_s\) and wave vector are independent of the channel width (when the latter is much larger than the Bohr radius), the amplitude of the charge density wave could not be pinpointed: its order of magnitude is clear, but its actual value appears to be strongly dependent on the width of the channel. This is most likely an artifact of our method of solution. A more accurate solution of the density profile, valid both near the center and at the edges of the channel, remains an interesting problem for future research.

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notice, however, that the compressibility changes sign in a discontinuous manner, jumping from $+\infty$ to $-\infty$, as the critical density is crossed.

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