Low-energy instability of flexural phonons in graphene

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We study the effect exerted by the electrons on the flexural phonons in graphene, accounting for the attractive interaction created by the exchange of electron-hole excitations. Combining the self-consistent computation of the phonon self-energy with renormalization group methods, we show that graphene has two different phases corresponding to soft and strong renormalization of the bending rigidity in the long-wavelength limit. In the first case, the system may have an intermediate scale in which the phonon dispersion is softened, but it manages finally to become increasingly rigid over large distance scales. The strongly renormalized phase is closer however to critical behavior, with an effective rigidity that becomes pinned in practice at very small values, implying a very large susceptibility for the development of a condensate of the flexural phonon field.

In recent years, the feasibility of fabricating graphene layers a single atom thick\textsuperscript{[1–3]} has led to a great deal of activity, because of their novel electronic properties and potential applications\textsuperscript{[4]}. Beyond its remarkable electronic transport attributes, it is becoming increasingly clear that graphene’s mechanical and electromechanical properties are just as remarkable. Despite it’s extreme thinness, it behaves as a stable metallic membrane with finite elastic moduli and rigidity. From this point of view, one of the most intriguing features of graphene is its tendency to develop ripples with a characteristic length scale. In exfoliated graphene, these ripples are correlated to some extent with the irregularities of the substrate on which the graphene sheet is deposited. But there is also evidence that they arise in part as an effect intrinsic to the two-dimensional membrane. When graphene was first fabricated, it came as a surprise that a purely two-dimensional material could exist, without being destabilized by large thermal or quantum fluctuations. From a theoretical point of view, it has been shown however that the coupling of flexural phonons to the in-plane vibrations in a crystalline membrane provides a mechanism to stabilize the two-dimensional system, leading to a finite critical temperature for the crumpling transition\textsuperscript{[7]}. The ripples are usually viewed then as a sort of imprint of the large fluctuations that should be present in the low-dimensional material\textsuperscript{[8]}. The ripples are expected to have a significant impact on the electronic transport in graphene\textsuperscript{[9]}. More generally, it has been shown that the scattering with flexural phonons may lead to a reduction of the lifetime of electron quasiparticles in graphene\textsuperscript{[10]}. On the other hand, the effect exerted by the electrons on the out-of-plane vibrations of a metallic membrane has been much less studied. There have been partial approaches in which the coupling of the charge density to the flexural phonons has been considered\textsuperscript{[11–13]}, but a zero temperature scaling analysis characterizing the possible critical behavior in the system has not been undertaken until now.

In this paper we investigate the many-body effects on flexural phonons in graphene, in a theory where these are coupled to the electron charge density as well as to the in-plane vibrations. These are known to give rise to a repulsive self-interaction between flexural phonons. We show that the exchange of electron-hole excitations leads to an interaction with the opposite character, although it plays in general the role of an irrelevant perturbation in the system. We will analyze the conditions in which this attraction may have a significant effect, finding that it leads in that case to a softening of the acoustic branch of out-of-plane phonons in graphene. The effective bending rigidity can be then strongly suppressed beyond a certain length scale, making the system very susceptible to the development of a condensate (nonvanishing average value) for the out-of-plane phonon field.

We characterize the shape of the graphene sheet by the vector field \( \mathbf{u} = (u_1, u_2, h) \), where \( u_1, u_2 \) represent the in-plane displacement with respect to the equilibrium position and \( h \) is the out-of-plane shift. The deformation of the membrane is analyzed in terms of the strain tensor \( u_{ij} \), given by

\[
u_{ij} = \frac{1}{2} \left[ \partial_i u_j + \partial_j u_i + \partial_i h \partial_j h \right]
\]

Thus, the action of the system has a term governing the dispersion of flexural phonons and depending on the mass density \( \rho \) and the rigidity \( \kappa \)

\[
S_{\text{free}} = \frac{1}{2} \int dt \, d^2x (\rho (\partial_t h)^2 - \kappa (\nabla^2 h)^2)
\]

while the dynamics of the in-plane phonons is controlled by a term of the form

\[
S_u = \frac{1}{2} \int dt \, d^2x (\rho (\partial_t u_i)^2 - 2\mu Tr u_{ij}^2 - \lambda (Tr u_{ij})^2)
\]

where \( \mu \) and \( \lambda \) are the Lamé coefficients.

The term \( S_u \) contains a quartic self-interaction for the out-of-plane phonon field \( h \). It is well-known that this interaction has to be taken into account for the description of stable crystalline membranes, as the soft dispersion in \[\text{[2]}\] leads to infrared divergences that cannot be tamed in the free theory\textsuperscript{[7]}. The novel effect arising in graphene is that the electronic charge couples to the displacement...
of the sheet, in such a way that it gives rise to another
source of self-interaction between the phonon fields. We
will consider the effect of electrons from the π bands of
graphene, represented by the field Ψ(x). Accounting for
the fact that the coupling must take place through the
trace of the strain tensor u_{ij}, we add to the action a new
term
\[ S_{e-ph} = -g \int dt d^2x \Psi^\dagger(x)\Psi(x) Tr u_{ij} \]  
(4)
The electron-phonon coupling g has dimensions of energy
and is directly related to the deformation potential of
the graphene lattice, which has a microscopic origin.

Assuming that the electron correlations are weak in
graphene, we may represent the dynamics of the Ψ(x)
field by a quadratic hamiltonian, matching at low ener-
gies the usual expression for the Dirac fermion fields. The
electron degrees of freedom can be integrated out at this
point, leading from (4) to a new phonon contribution to the
action, to be added to (2) and (3),
\[ S_u = -\frac{1}{2}g^2 \int d\omega_q d^2q \chi(q, \omega_q)(Tr u_{ij})(Tr u_{ij}) \]  
(5)
where \( \chi(q, \omega_q) \) is the density-density correlator depend-
ing on frequency and momentum variables. Terms that
are higher order in \( Tr u_{ij} \) can be generated after inte-
gration of the electron fields, but they turn out to be
more irrelevant perturbations in the approach developed
in what follows.

Since the energy scale of electronic excitations (for a
given momentum) are much larger than the phonon en-
geries, we can safely take the limit \( \omega = 0 \) in the charge
susceptibility. In the Dirac theory, we have in particular
\( \chi(q, 0) = -|q|/v_F \). We observe that the new contribu-
tion \( S_u \) tends to cancel the term with the LamÉ co-
efficient \( \lambda \) in (3). After following the usual procedure to
integrate the in-plane phonons \( u_\parallel \) [4], we get the total
action for the flexural phonons
\[ S = \frac{1}{2} \int d\omega_q d^2q (\rho \omega_q^2 h^2 - \kappa_q^4 h^2) \]
\[ -\frac{1}{2} \int d\omega_q d^2q K(q)\bar{u}_{ij}(q, \omega_q)u_{ij}(-q, -\omega_q) \]  
(6)
where \( \bar{u}_{ij}(q, \omega_q) \) is the Fourier transform of
\( (1/2)P_{ij}\partial_\parallel \partial_\parallel h, \) \( P_{ij} \) being the transverse projector,
and the quartic coupling is (in the Dirac theory)
\[ K(q) = \frac{4\mu(\mu + \lambda - g^2|q|/v_F)}{2\mu + \lambda - g^2|q|/v_F} \]  
(7)

Negative values of the coupling function \( K(q) \) may lead
to an instability of the graphene sheet, as they represent
an attractive interaction between the flexural phonons.
The phonon self-energy and the interaction vertices are
actually affected by logarithmic divergences in the energy
cutoff, which makes it necessary to keep track of the low-
energy dependence of the couplings. In order to study
the competition between positive and negative couplings
in (7), we may approximate this coupling function by the
constant term \( K_0 = 4\mu(\mu + \lambda)/(2\mu + \lambda) \) and the dominant
powers of \( |q| \)
\[ K(q) \approx K_0 - G_1 |q| - G_2 q^2 \]  
(8)
Neglecting higher-order terms in \( |q| \) amounts to disre-
garding the exchange of an increasing number of electron-
hole pairs in the in-plane phonon propagator, which is
justified as long as the pole arising from the denomina-
tor of (7) is not included within the first Brillouin zone of
a real graphene system. This is consistent with an esti-
mate for the on-site deformation potential for the in-
plane phonons 5\( \mu_0 \approx 8 \) eV [14], that is the coupling ap-
pearing in the denominator of Eq. (7).

The different interactions tend to modify the low-
energy behavior of the rigidity \( \kappa \) in opposite directions
depending on their attractive or repulsive character. This
can be seen through the corrections to the phonon self-
energy \( \Sigma(p, \omega_p) \) represented in Fig. 1. Computing
the full propagator of the flexural phonons from the expres-
sion \( D^{-1}(p, \omega_p) = \omega_p^2 - \left(\kappa_0/\rho\right)p^2 - \Sigma(p, \omega_p) \), we find the
renormalized bending rigidity \( \kappa(p) \) given by the equa-
tion
\[ \frac{\kappa(p)}{\rho} = -\frac{\kappa_0}{\rho} + \frac{1}{8\pi^2} \int_0^{q_0} dq \int_0^{2\pi} d\phi \sin^4(\phi) \frac{K_0 - G_1 |p - q| - G_2 |p - q|^2}{\rho^2 |p - q|^4} \]  
(9)
A self-consistent treatment of the many-body theory re-
quires that the rigidity arising in the integrand of (10)
from the phonon propagator must be also taken as \( \kappa(p) \).
This leads to an integral equation, that can be solved to
obtain the effective momentum dependence of the renor-
malized parameter.

We have represented in Fig. 2(a) the result of solving
Eq. (9) in different regimes of the coupling \( g \). We ob-
serve that, for a sufficiently large value \( g_0 \) , the function
\( \kappa(p) \) vanishes at a certain momentum, bouncing back for
smaller values of \( |p| \). In this picture, \( g_0 \) plays the role of
critical coupling, as larger values of \( g \) would actually lead
to negative values of the bending rigidity, implying the
absence of a self-consistent solution. In that case, how-
ever, a more refined analysis has to be applied in order
\[ \text{FIG. 1. First order corrections to the self-energy of flexural}
\text{phonons (represented by a wiggly line) arising from (a) the}
\text{four-phonon interaction } K_0 \text{ and (b) the exchange of electron-}
\text{hole excitations (represented by the bubble with arrow lines).} \]
The main scaling dependence is given by the behavior
\[ G_n(q) = q^n \tilde{G}_n(q) \]  
where the couplings \( \tilde{G}_n \) are subject to a purely logarithmic scaling in the same fashion as \( K_0 \).

A scaling equation can be also written for the rigidity \( \kappa \), as the self-energy diagrams in Fig. 1 actually display a logarithmic dependence on the cutoff \( q_c \). Taking into account the irrelevant character of the couplings \( G_n \), we find that under a variation of the momentum scale \( q \)

\[
\frac{q}{\rho} \frac{\partial \kappa}{\partial q} = -\frac{3}{16\pi} \frac{K_0/\rho^2}{(\kappa/\rho)^{3/2}} + \frac{3}{16\pi} \sum_n q^n \frac{G_n/\rho^2}{(\kappa/\rho)^{3/2}}
\]

Eq. (13) has to be solved then together with (10) and (11) in order to obtain the behavior of the effective rigidity \( \kappa \). We note that, in real graphene samples, the bare values of the couplings \( K_0 \) and \( G_n \) place in general the system in the weak-coupling regime. On the one hand, the energy scale of \( K_0 \) is given by \( a^2/4\pi(\mu + \lambda)/(2\mu + \lambda) \approx 20 \text{ eV} \), where \( a = 1.4 \text{ Å} \) is the interatomic spacing. The coupling from the electronic sector is subject to the uncertainty in the estimates of the deformation potential for out-of-plane vibrations, leading to a wide range of values with \( q \sim 20 - 30 \text{ eV} \). On the other hand, we have the energy scale \( a^2/\pi(\kappa/\rho)^{3/2} \), which has a nominal value of \( \approx 40 \text{ eV} \) in graphene. When \( \kappa/\rho \) becomes very small, however, the rate of decrease of \( K_0, G_1 \) and \( G_2 \) is greatly increased, leading to significant differences with respect to the behavior of the effective rigidity obtained from the self-consistent equation (10).

The behavior of \( \kappa \) obtained from the scaling equations is shown in Fig. 2(b) for different bare values of \( g \). For small values of this coupling, we see that the effective rigidity has an essentially increasing trend as \( q \to 0 \). There is however a regime of large \( g \), corresponding to values of the deformation potential \( q \sim 25 - 30 \text{ eV} \), in which \( \kappa \) is significantly reduced due to the electron-phonon interaction. At some point, \( \kappa \) may be even suppressed by several orders of magnitude, but without vanishing as in the self-consistent approach shown above. Here we find instead that \( \kappa \) always bounces back from zero, though it may become trapped in a range of very small values as the effective couplings \( K_0, G_1 \) and \( G_2 \) are themselves quickly renormalized very close to zero.

We find therefore that graphene has two different phases corresponding to soft and strong renormalization of \( \kappa \) as \( q \to 0 \). In the first case, the system may have an intermediate scale in which the effective \( \kappa \) is reduced, but it manages finally to become increasingly rigid over large distance scales. In the strongly renormalized phase, however, the effective rigidity becomes pinned in practice at very small values below a certain momentum scale.

Although the self-consistent solutions of Eq. (10) miss the renormalization of the couplings at very small \( \kappa \), we can still use them to draw the boundary between the phases with soft and strong renormalization of the bending rigidity. That line can be formed with the values of critical coupling \( g_c \), beyond which the integral equation would lead to negative values of \( \kappa \). This forbidden region corresponds in the renormalization group approach to the

\[
\frac{q}{\rho} \frac{\partial \kappa}{\partial q} = -\frac{3}{16\pi} \frac{K_0/\rho^2}{(\kappa/\rho)^{3/2}} + \frac{3}{16\pi} \sum_n q^n \frac{G_n/\rho^2}{(\kappa/\rho)^{3/2}}
\]

For \( \kappa \leq 0 \), this leads to a negative value of \( q/\rho \) and hence the integral equation may not be solved. The critical coupling \( g_c \) is given by the condition \( \kappa = 0 \), and we find

\[
q = \frac{3}{16\pi} \frac{K_0/\rho^2}{(\kappa/\rho)^{3/2}}
\]

This critical coupling is shown in Fig. 2(b) for different bare values of \( g \). The phase diagram of graphene is shown in Fig. 2(b) for different bare values of \( g \). The phase diagram shows two different phases corresponding to soft and strong renormalization of \( \kappa \) as \( q \to 0 \). In the first case, the system may have an intermediate scale in which the effective \( \kappa \) is reduced, but it manages finally to become increasingly rigid over large distance scales. In the strongly renormalized phase, however, the effective rigidity becomes pinned in practice at very small values below a certain momentum scale.
FIG. 3. Diagram showing the phases corresponding to a rigid long-distance regime of graphene (lower region) and pinning of the renormalized bending rigidity at very small values (upper region), in the space of nominal values of the rigidity $\kappa_0$ and deformation potential $g$. The value of $a^2K_0$ was taken as 18 eV.

The phase with strong renormalization of the bending rigidity can be put in correspondence with the appearance of ripples in graphene, as very small values of $\kappa$ imply a very large susceptibility for the development of a condensate (nonvanishing average value) of the flexural phonon field $h$. It is conceivable that the different degree of Coulomb screening may control which of the two phases is found in a real graphene sample, as the Coulomb interaction leads to a slight reduction in the charge susceptibility. Thus, graphene can be more prone to develop ripples or to buckle when surrounded by media with high dielectric constant, for which the values of $\chi(q,0)$ are maximized.

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