Thermal fluctuations of crystalline membranes and ripple formation in graphene

N. Hasselmann

Crystalline membranes are two-dimensional membranes whose constituent particles form a crystalline mesh. A good example is free standing graphene where carbon atoms form a membrane with a hexagonal lattice structure. What distinguishes crystalline membranes from conventional two-dimensional crystals is the embedding in a three dimensional space which allows for fluctuations not only within the two internal dimensions of the membrane (in-plane modes) but also for fluctuations in the direction perpendicular to the plane of the membrane (out-of-plane or flexural modes) [1,2]. As in conventional crystals, crystalline membranes have, at least at small length scales, a finite shear modulus besides a finite bulk modulus.

The coupling of in-plane and out-of-plane fluctuations gives rise to an exotic elasticity of the membrane. In the infrared limit, the elastic properties are highly unusual. These include the absence of any finite elastic constants in the thermodynamic limit, a negative Poisson's ratio (i.e., if the membrane is compressed in one direction, it also shrinks in the other direction), and fluctuations characterized by a large anomalous dimension. At high temperatures the fluctuations become eventually so large that the membrane undergoes a crumpling transition from a so-called flat phase, in which the average direction of the vector normal to the membrane is non-vanishing (i.e. the membrane spans a well-defined two-dimensional plane), to a crumpled state in which the normals are not ordered. Despite more than two decades of research, there is still no consensus on the nature of the crumpling transition.

Free standing graphene, which is a mono-atomic two-dimensional crystal, should be an ideal crystalline membrane. On the theoretical side, a negative Poisson ratio and anomalous fluctuations have been seen in Monte Carlo (MC) simulations based on an realistic effective many-body interaction of carbon atoms. While anomalous fluctuations have not yet been seen in experiments, they have been observed in simulations [3]. One of the most surprising experimental outcome of experimental investigations of free standing graphene were the observation of ripples in graphene sheets with a characteristic scale $50 - 100\,\text{Å}$, which where also reproduced in MC simulations. Ripple formation has often been argued to have its origin in the coupling of the elastic modes (phonons) to the electronic degrees of freedom which are gapless Dirac quasiparticles. Here we argue instead that ripples are a direct result of the nonlinear elasticity of the membrane and their presence should be viewed as a confirmation of the theory of crystalline membranes [1].

Since we want to analyse the behavior of the membrane not just at very small momenta but also at intermediate scales relevant for ripples, we analyse the membrane fluctuations also beyond the small momentum regime $q \to 0$ where they become anomalous. For this, we use a nonperturbative renormalization group (NPRG) approach [1,2]. Perturbative RG approaches, such as field theoretical RG, rely on an expansion in a small parameter. Such a parameter is however not readily available for physical crystalline membranes. Different perturbative approaches, based e.g. on a ε-expansion where the difference of the internal dimensionality D of the membrane and the upper critical dimension (which is four) is the expansion parameter, or a $1/d$ expansion where d is the dimension of the embedding space, have produced conflicting results.

We describe the in-plane modes by standard elasticity theory which is, to lowest order in the elasticity tensor, characterized by the two Lamé constants μ and λ, with the bulk modulus given by $\mu + \lambda$ and the shear modulus by μ. The embedding into three-dimensional space gives rise to a bending energy which is described by an additional bending constant κ. Starting point of our analysis is a commonly used Landau-Ginzburg ansatz for the energy functional of crystalline membranes,

$$\mathcal{H} = \frac{\tilde{\kappa}}{2} \int d^2x \, (\partial_a \partial_a \boldsymbol{R})^2 + \int d^2x \Big[\frac{\tilde{r}_0}{2} (\partial_a \boldsymbol{R})^2 + \frac{\tilde{\mu}}{4} (\partial_a \boldsymbol{R} \cdot \partial_b \boldsymbol{R})^2 + \frac{\tilde{\lambda}}{8} (\partial_a \boldsymbol{R} \cdot \partial_a \boldsymbol{R})^2 \Big] , \tag{1}$$

where the first term, proportional to the bending constant $\tilde{\kappa} = \kappa/(k_B T)$, describes the bending energy and the remaining terms describe the stretching part with $\tilde{\mu} = \mu/(k_B T)$ and $\tilde{\lambda} = \lambda/(k_B T)$. Here, $\boldsymbol{R}(\boldsymbol{x})$ is a $2+1$ dimensional vector parameterizing the 2 dimensional membrane which is embedded in a $2+1$ dimensional space, see Fig. 1. The presence of an UV cutoff, which arises from the underlying lattice structure, is implicitly assumed. The model (1) can be tuned through the crumpling transition by varying the parameter $\tilde{r}_0$. The flat phase is characterized by a finite order parameter $\boldsymbol{m}_{a,0} := \langle \partial_a \boldsymbol{R} \rangle \neq 0$ of magnitude $J = |\boldsymbol{m}_{a,0}|$. For positive values of $\tilde{r}_0$ a crumpled state is realized with no broken symmetries.

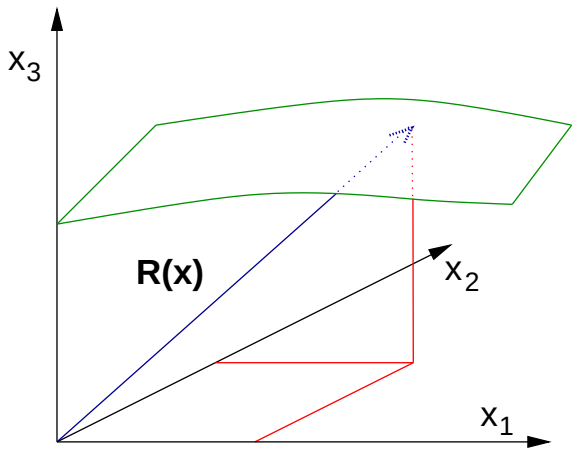

Figure 1: Parameterization $\boldsymbol{R}(\boldsymbol{x})$ of the membrane. The mapping $\boldsymbol{R}(\boldsymbol{x})$ is from the two-dimensional vector $\boldsymbol{x} = (x_1, x_2)$ to the three-dimensional embedding space of the membrane (membrane shown with green contours).

The NPRG approach is based on an exact flow equation for the cutoff dependent effective average action Γ_Λ, which yields the fully renormalized irreducible vertices in the limit of $\Lambda \to 0$. Initially, $\Gamma_{\Lambda_0} = \mathcal{H}$, but for $\Lambda < \Lambda_0$ the coupling constants $\tilde{\kappa}$, $\tilde{\mu}$ and $\tilde{\lambda}$ will aquire a momentum dependence. We thus write the ansatz [1,2]

$$\begin{aligned}\Gamma_\Lambda &= \frac{1}{2}\int d^D x\, d^D x' \tilde{\kappa}_\Lambda(\boldsymbol{x}-\boldsymbol{x}')\partial_a^2 \boldsymbol{R}(\boldsymbol{x})\partial_b^2 \boldsymbol{R}(\boldsymbol{x}') \\ &\quad + \int d^D x\, d^D x' \Big[\tilde{\mu}_\Lambda(\boldsymbol{x}-\boldsymbol{x}')U_{ab}(\boldsymbol{x})U_{ab}(\boldsymbol{x}') + \frac{1}{2}\tilde{\lambda}_\Lambda(\boldsymbol{x}-\boldsymbol{x}')U_{aa}(\boldsymbol{x})U_{bb}(\boldsymbol{x}')\Big], \end{aligned} \tag{2}$$

where $U_{ab} = (1/2)(\partial_a \boldsymbol{R}\cdot\partial_b \boldsymbol{R} - \boldsymbol{m}_{a,0}\cdot\boldsymbol{m}_{b,0})$ is the stress tensor. The advantage of this ansatz is that all relevant symmetries of the model are explicitly obeyed. Furthermore, the flow equations corresponding to the ansatz (2) are unique and can be solved numerically, without any further approximation. In Fig. 2 we show the complete momentum dependence for the out-of-plane fluctuations $G_{hh}(q) = \langle |h_q|^2 \rangle$ where h are the out-of-plane distortions of the membrane. We choose $T = 300$ K, with initial values for the elastic constants appropriate for graphene taken from previous MC investigations. The data from MC simulations [3], based on a highly realistic carbon-carbon interaction potential, are also shown. It is obvious that the agreement is excellent throughout the entire momentum regime and deviations appear only close to the Bragg momentum.

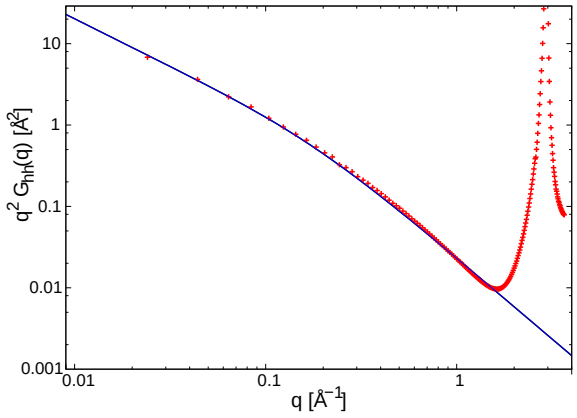

Figure 2: Results for the out-of-plane fluctuations $q^2 G_{hh}(q)$ from NPRG (solid, black) [1] and MC simulations [3] (dashed, red). A crossover from the perturbative q^{-4} scaling at large momenta to the anomalous scaling $q^{-4+\eta}$ in the small momentum regime is clearly visible. The crossover momentum is $q_G \approx 0.08\,\text{Å}^{-1}$. The large peak in the MC data corresponds to a Bragg vector of the lattice.

For large momenta, the out-of-scale fluctuations follow the q^{-4} behavior of the non-interacting theory, whereas for very small momenta the fluctuations are strongly renormalized and behave as $q^{-4+\eta}$ with a large anomalous dimension $\eta \approx 0.85$, a value also consistent with the MC simulations. Both the anomalous regime and the perturbative regime are characterized by an algebraic decay of correlations and do not reveal any characteristic length. However, the crossover between these two regimes occurs at a finite Ginzburg or crossover scale q_G which we find to be $q_G \approx 0.08\,\text{Å}^{-1}$. This corresponds to a real space length scale of about $80\,$Å, which is just the characteristic scale of ripples which have been observed both in MC simulations and in experiments. This therefore suggests that ripples in free standing graphene have their origin in the nonlinearity of the elastic theory of crystalline membranes. Moreover, a perturbative analysis of the crossover scale reveals that it is approximately given by $q_G \propto (K_0 k_B T)^{1/2}/\kappa$, where K_0 is the (bare) Young's modulus $K_0 = 4\mu(\mu+\lambda)/(2\mu+\lambda)$. Thus, q_G scales with temperature as $q_G \approx \sqrt{k_B T}$, assuming that the bare elastic constants do not change with temperature.

The NPRG approach can also be used to investigate the nature of the crumpling transistion at which the mem-

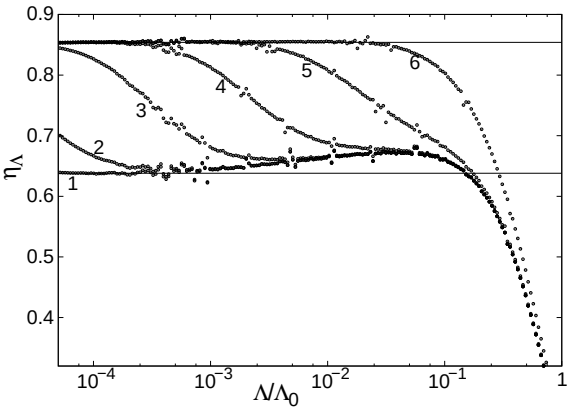

Figure 3: RG flow of the anomalous dimension as a function of the cutoff Λ, for different initial values of the parameter $\tilde{r}_0$ (labels 1-6). There are two different fixed points. The one with $\eta \simeq 0.85$ corresponds to the flat phase whereas the one with $\eta \simeq 0.63(8)$ corresponds to the crumpling transition.

brane loses the orientational order of its normals. Note that the crumpling transition plays no role for graphene since the graphene layer would be destroyed at temperatures much smaller than the crumpling transition. The transition is however an important issue in statistical mechanics and it is still not well understood. It is not even clearly established whether the transition is of (weakly) first or of second order. All previous RG approaches to the transition have dealt only with a finite number of coupling constants. Our NPRG ansatz, which includes infinitely many coupling terms, is thus expected to yield more reliable results.

In our numerical analysis of the NPRG flow equations we find, besides the fixed point of the flat phase, another fixed point at which the order parameter J vanishes and which describes the vicinity of the crumpling transition [2], see Fig. 3. The presence of this fixed point is a signature of a second order phase transition. We find an anomalous exponent $\eta_{cr} \approx 0.63(8)$, whereas the order parameter vanishes with the critical exponent $\beta \approx 0.22$. We further determine the thermal exponent $\nu \approx 0.69$ and the Poisson's ratio at the transition is $\sigma \approx -0.71(5)$, rather close to the smallest possible value -1. The values of the critical exponents are in reasonable agreement with those obtained from simulations where a second order transition is found.

In summary, we investigated for the first time the full momentum dependence of crystalline membranes, using a nonperturbative renormalization group approach. We find a second order crumpling transition and determined the critical exponents. Choosing bare values of the elastic coupling constants appropriate for graphene, we further find excellent agreement with MC simulations. The out-of-plane fluctuations are characteristed by a single crossover scale which coincides with the rippling scale of free standing graphene. Ripples are thus interpreted as a direct manifestation of the nonlinear nature of the membrane elasticity, which would make them precursors of anomalous fluctuations which are expected at smaller momenta.

References:

[1] *Braghin, F. L. and N. Hasselmann*, Physical Review B **82**, 035407 (2010).

[2] *Hasselmann, N. and F. L. Braghin*, Physical Review E **83**, 031137 (2011).

[3] *Fasolino, A., J. H. Los, and M. I. Katsnelson*, Nature Materials **6**, 858-861 (2007); *Los, J. H. , M. I. Katsnelson, O. V. Yazyev, K. V. Zakharchenko, and A. Fasolino*, Physical Review B **80**, 121405(R) (2009).

In collaboration with:

F. L. Braghin (Univ. Fed. de Goiás, Goiânia, Brazil)