Lindemann Parameters for solid Membranes focused on Carbon Nanotubes

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Temperature fluctuations in the normal direction of planar crystals such as graphene are quite violent and may be expected to influence strongly their melting properties. In particular, they will modify the Lindemann melting criterium. We calculate this modification in a self-consistent Born approximation. The result is applied to graphene and its wrapped version represented by single-walled carbon nanotubes (SWNTs). It is found that the out-of-plane fluctuations dominate over the in-plane fluctuations. This makes strong restrictions to possible Lindemann parameters. Astonishingly we find that these large out-of-plane fluctuations have only a small influence upon the melting temperature.

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I. INTRODUCTION

The production of macroscopic two-dimensional (2D) graphene sheets by mechanical cleaving \(^1\) has demonstrated that free-standing or suspended 2D crystals can exist despite large positional fluctuations in two dimensions. Since then, a variety of other free-standing 2D crystallites have been prepared \(^2\). Wrapped versions of the 2D free-standing graphene had been found much earlier in 1991 \(^3\). Recent observations \(^4\) have confirmed the theoretical expectation that freely suspended graphene sheets are strongly undulated and behave more like solid membranes than two dimensional (2D) crystals \(^3\). The undulations are a consequence of the thermal fluctuations of the membrane. In this paper, we calculate these fluctuations quantitatively and discuss their implications upon the melting properties such as Lindemann parameter and melting temperature. \(^4\) The results will be compared with corresponding 2D crystals.

The easiest way to estimate the melting temperature of a three dimensional (3D) crystal is based on the Lindemann criterium \(^5\). According to it, a 3D crystal starts to melt when the square root of the thermal expectation value of lattice site elongations \(\sigma_i \equiv \sqrt{\langle u_i^2 \rangle_T} \) exceeds a certain fraction of the nearest-neighbor lattice distance \(a\), usually around 0.1–0.15 \(^7\). Above the melting temperature, the shear modulus of the lattice vanishes leading to a divergence in the displacement fluctuations typical for the liquid state.

In two dimensions (2D) this criterium is no longer applicable since the displacement fluctuations are always logarithmically divergent, reflecting the fact that after a long time, a 2D crystal migrates through the entire 2D-space. There exists, however, a simple modification \(^8\). Instead of \(\sqrt{\langle u_i^2 \rangle_T} \) one may use the finite cumulants \(\langle r_{ij}^2 \rangle_T - \langle r_{ij} \rangle_T^2 \), where \(r_{ij}\) is the difference vector between the atoms associated with the nearest-neighbor lattice sites \(i\) and \(j\). This leads to a modified Lindemann number

\[
\mathcal{L}^*_{1,2D} = \frac{1}{|N_1|} \sum_{i,j \in N_1} \sqrt{\langle r_{ij}^2 \rangle_T - \langle r_{ij} \rangle_T^2} / a. \tag{1}
\]

Here \(N_1\) denotes the set of all nearest-neighbor lattice pairs and \(|N_1|\) their number. For the Lennard-Jones and Wigner lattices, Bedanov et al. \(^8\) found by computer simulations values of \(\mathcal{L}^*_{1,2D} \approx 0.15 - 0.2\). We have derived the same values analytically for a triangular generalization \(^9\) of a square lattice defect melting model \(^7\).

At this point it is useful to realize that a migration problem and an associated divergence of \(\sigma_i\) exists also in three dimensions if the system is finite, i.e., for 3D clusters and polymers. There one defines a modified Lindemann number

\[
\mathcal{L}^*_{1,3D} = \frac{1}{|N_{1,2}|} \sum_{i,j \in N_{1,2}} \sqrt{\langle r_{ij}^2 \rangle_T - \langle r_{ij} \rangle_T^2} / \langle r_{ij} \rangle_T. \tag{2}
\]

Here \(N_{1,2}\) is the set of all lattice site pairs whose number is \(N(N-1)/2\) where \(N\) is the number of atoms in the lattice. The number \(\mathcal{L}^*_{1}\) was introduced by Kaelberer and Etters \(^10\), the number \(\mathcal{L}^*_{2}\) by Berry et al. \(^11\). For small clusters, \(\mathcal{L}^*_{1}\) and \(\mathcal{L}^*_{2}\) have similar values \(^12\) of around \(\mathcal{L}^*_{1,2} \approx 0.03 - 0.05\) at the melting point. Above the melting point all modified Lindemann numbers increase considerably, but do not go to infinity (this being in contrast to \(\sigma_i\)).

The main difference between \(\mathcal{L}^*_{1,3D}\) and \(\mathcal{L}^*_{2}\) comes from the last term in the square root in \(^1\) and \(^2\). Whereas \(\langle r_{ij} \rangle_T\) is the temperature average of the difference vector of sites \(i\) and \(j\), i.e. \(r_{ij} = (x_{ij}, y_{ij}, z_{ij})\), the expectation value \(\langle r_{ij} \rangle_T\) is the average value of the bonding length of sites \(i\) and \(j\), i.e. \(r_{ij} = (x_{ij}^2 + y_{ij}^2 + z_{ij}^2)^{1/2}\). Since \(r_{ij}^2 = r_{ij}^2\) one would expect that the 3D version of \(^1\), \(\mathcal{L}^*_{1,3D}\),...
and $\mathcal{L}_1^s$ could be equally useful in determining the melting point. This is indeed the case for 3D crystals.

In this paper we shall consider all three Lindemann numbers $\mathcal{L}_1^{s,3D}$, $\mathcal{L}_c^s$, and $\mathcal{L}_c^2$ as candidates for a melting criterion for solid membranes such as graphene lattices or SWNTs. It will turn out that for these $\mathcal{L}_1^{s,3D}$ is unsuitable for calculating the melting temperature. The reason lies in the large out-of-plane fluctuations of the membrane varying little when crossing the melting point. These fluctuations cancel each other in (2) since $\langle z_{ij}^2 \rangle_T \neq 0$ and $r_{ij} \approx (x_{ij}^2 + y_{ij}^2)^{1/2} + (1/2)z_{ij}^2/(x_{ij}^2 + y_{ij}^2)^{1/2}$ but not in since $\mathcal{L}_1^{s,3D}$ since $\langle z_{ij} \rangle_T = 0$.

Freely suspended graphene sheets are always undulated and behave like a solid membrane [3]. Nelson et al. [3] have shown that in-plane fluctuations tend to stabilize a solid membrane such that a flat phase can exist in spite of its large 2D fluctuations. The melting temperature of (5,5) SWNTs was determined by Zhang et al. [14] within numerical simulation to be around $T_m \approx 5000$ K, in agreement with experimental determinations [15]. The value of the Lindemann number $\mathcal{L}_c^s$ was around $\mathcal{L}_c^s \approx 0.03$ at the onset of melting defined by the abrupt increase of $\mathcal{L}_c^s$. However, when using the region of strong increase of the internal energy they obtain a range $\mathcal{L}_c^s \approx 0.03 \sim 0.07$ from the onset of melting to its completion.

The shapes of SWNTs near the melting temperature are in general strongly deformed from a pure tube form. This leads to the conclusion that the 2D nearest-neighbor Lindemann number $\mathcal{L}_1^{s,2D}$ [11] is not a useful quantity for a melting criterion. One rather should use the Lindemann-like numbers [2] or the 3D form of [11] which both respect the 3D rotational symmetry of the system. In the following, we shall first calculate $\mathcal{L}_1^s$ which for small clusters and small supercells agrees in molecular dynamic simulations with $\mathcal{L}_c^s$. We shall restrict ourselves to the (5,5) SWNT so that we can compare our theoretical results with existing simulation data. We shall find that despite the large vertical fluctuations of the membranes, the Lindemann number [2] depends mostly on the in-plane fluctuations and provides us with a valuable melting criterion. This is not the case for the 3D-version of the Lindemann number $\mathcal{L}_1^s$ having its reason in the fact that the out-of-plane fluctuations are even larger than the in-plane fluctuations for SWNTs and graphene. Surprisingly, the melting temperature of SWNTs is modified only little by these large out-of-plane fluctuations at high temperatures.

II. MEMBRANES

The elastic energy of a solid elastic membrane in the flat phase is given by [16]

$$H_{el} = \int d^2x \left[ \mu u_{ij}^2 + \frac{1}{2} \lambda u_{ij}^2 + \sigma_{ij} u_{ij} + \frac{1}{2} \kappa_0 (\nabla_i \nabla_j f)^2 \right] ,$$

where

$$u_{ij} = \frac{1}{2} (\nabla_i u_j + \nabla_j u_i + \nabla_i f \nabla_j f) .$$

and $u_{ij}$ are the lattice displacements in $xy$-plane, while $f$ is the out-of-plane displacement. The constant $\mu$ is the shear modulus and $\lambda$ is the Lamé constant. The last term in (4) with the constant $\kappa_0$ accounts for the bending stiffness of the membrane. The quantity $\sigma_{ij}$ is an external stress source which will help us to calculate (1,2,4) from derivatives of the partition function with respect to $\sigma_{ij}$. The line element on the membrane for small distortions is given by [16] $d l^2 = d l^2 + 2 u_{ij} d x_i d x_j$, where $d l$ is the length of the undistorted planar surface, which we identify with the equilibrium lengths $l_{ij}$ between sites $i$ and $j$. Thus we calculate $\mathcal{L}_{2,1}^c$ by first inserting the line element $d l'$ in (2), and afterwards expanding the resulting expressions for small displacements. Thus we obtain

$$\mathcal{L}_{2,1}^c = \sum_{i,j \in \Omega_{1,2}} \frac{\langle u_{lm} a e^T e l m \rangle^2_T}{\langle u_{lm} a e^T e l m \rangle^2} .$$

Here $e^T$ are the unit vectors pointing from site $i$ to $j$.

In deriving (5) we used a Taylor expansion of the elongation differences between two lattice sites (gradient expansion). This is justified for the small elongation differences of neighboring atoms occurring in $\mathcal{L}_1^s$ and $\mathcal{L}_c^s$ up to the melting regime. In the atom pairs summed over in $\mathcal{L}_2^s$, the approximation is good only for small clusters or for small supercells in molecular dynamic simulations. For infinite solid membranes, this is no longer the case. The contributions of the far separate pairs $(i, j)$ cause a drastic decrease of the Lindemann parameter $\mathcal{L}_c^s$ with the size of the system. In a 2D crystal, for example, the widely separated pairs contribute terms which grow logarithmically with the separation: $\lim_{l_{ij} \to \infty} (r_{ij}^3/T) - (r_{ij}^3)^2 \sim \ln l_{ij}$. This eliminates $\mathcal{L}_c^s$ for determining the melting point.

In order to calculate (4) we first integrate out the $xy$-lattice displacement fields $u_{ij}$ in the partition function, leading to an effective Hamiltonian $H = H_h + H_{\sigma \sigma}$ with

$$H_h = \int d^2x \left\{ \tilde{\mu} (h_f + h_{\sigma})^2 - \frac{1}{2} \kappa_0 (\nabla_i \nabla_j f)^2 \right\} ,$$

$$H_{\sigma \sigma} = \int d^2x \left[ \frac{1 - \nu}{4\mu} (\nabla_i \nabla_j \sigma_{ij})^2 + \frac{1}{2\mu} (\nabla_i \nabla_j \sigma_{ij}^T)^2 \right] ,$$

and the energy densities

$$h_f = \frac{1}{2} P_{lm}^T (\nabla_i f \nabla_m f) ,$$

$$h_{\sigma} = \frac{1}{2\mu} P_{lm}^T \left[ 1 - \delta_{lm} \frac{\nu}{1 + \nu} \left( 1 + \frac{\nabla_i \nabla_j}{1 - \delta_{ij} \kappa_0 \kappa_0} \right) \right] .$$

In (6) we have used the abbreviation $\tilde{\mu} \equiv \mu (1 + \nu) = E/2$ where $E$ is the Young modulus and $\nu \equiv \lambda/(2\mu + \lambda)$ the Poisson ratio. The calculation of the energy densities
is simplified by the fact that only the transverse part \( P_{lm}^T \nabla f \nabla m \) with \( P_{lm}^T = (\delta_{lm} - q t_q m / q^2) \) of the out-of-plane fluctuations is relevant after integrating out the in-plane fields \( u_i \). The transverse projections lead to a useful restriction of the relevant phase space when calculating Feynman diagrams.

### A. Self-consistent Born approximation

We now treat the Hamiltonian \( \mathcal{H} \) within the self-consistent Born-approximation (SCBA) corresponding to the Hartree-Fock approximation for the eigenfunctions. Other approximations to the Hamiltonian \( \mathcal{H} \) have been used \( \text{[17]} \) to calculate the universal roughening exponents of the membrane, for example in Ref. \( \text{[18]} \) an extension of SCBA.

Denoting the inverse Green function of the \( f \)-fluctuations by \( G^{-1}(k) = \kappa_0 k^4 + \Sigma(k) \), we obtain from \( \text{[19]} \) within the SCBA

\[
\Sigma(k) = \frac{2 \mu k_B T}{(2\pi)^2} \int_{BZ} d^2 q \frac{(k \times q)^4}{q^4} G(k + q)
\]

where we take into account only the Fock-part of the SCBA. It was shown in Ref. \( \text{[13]} \) that the Hartree-terms do not contribute for free boundary conditions of the \( xy \)-elongations of the membrane. To do the integral \( \text{[20]} \) we use a circular Brillouin zone \( k \leq k_{BZ} \), with \( k_{BZ} = 8\pi / \sqrt{a^2} \) for the triangular Bravais lattice of SWNTs and graphene. The integral \( \text{[20]} \) can be carried out exactly for small \( k \) \( \text{[19]} \) to obtain the first two terms in the expansion

\[
\Sigma(k) = C_T k^3 + \kappa_T k^4 + \ldots,
\]

where \( C_T \) is a temperature-dependent constant which turns out to be

\[
\frac{C_T}{\kappa_0 k_{BZ}} = \sqrt{T/2\pi}.
\]

The symbol \( \bar{T} \) denotes the dimensionless temperature \( \bar{T} = \mu k_B T / (\kappa_0 k_{BZ})^2 \).

The second coefficient \( \kappa_T \) is determined as follows. We assume that the truncated small-\( k \) expansion \( \text{[21]} \) can be used for all \( k \) in the Brillouin zone, implying that the inverse Green function has the form

\[
G^{-1}(k) \approx C_T k^3 + \kappa_T k^4,
\]

with \( \kappa_T \equiv \kappa_0 + \kappa_T \). We shall see below that this assumption is indeed justified. At low temperature where \( C_T \ll \kappa_0 k_{BZ} \) we determine \( \kappa_T \) by inserting \( \text{[21]} \) into \( \text{[20]} \) and evaluating the integral for \( \Sigma(k) \) at the momentum \( k = C_T / \kappa_T \). This momentum regime is most relevant in the integrals over \( G \) which we have to calculate in the following in order to determine the generalized Lindemann parameters. Moreover, we will show below that \( \text{[20]} \) is then justified in good approximation for momenta even in the whole Brillouin zone. At higher temperatures where \( C_T \gg \kappa_0 k_{BZ} \), we determine \( \kappa_T \) by integrating \( \text{[22]} \) at \( k = k_{BZ} \). In both temperature regimes we carry the momentum integrations up to \( k = k_{BZ} \), and obtain in either case a quadratic equation for \( \kappa_T \), solved by

\[
\frac{\kappa_T}{\kappa_0} \approx \begin{cases} \frac{3\bar{T}}{8\pi} \left( 1 - \sqrt{1 - \frac{15}{16\pi} \bar{T}} \right)^{-1} & \text{for } C_T \ll \kappa_0 k_{BZ}, \\ 1 - \frac{3}{4\sqrt{2\pi}} \sqrt{\bar{T}} & \text{for } C_T \gg \kappa_0 k_{BZ}. \end{cases}
\]

(12)

Our approximations are justified in Fig. 1 showing in the main plot the quantity \( G^{-1}(k) = \kappa_T k^4 + C_T k^3 \) divided by the sum of \( \kappa_0 k^4 \) and the numerically integrated right-hand side of the self-energy function \( \Sigma(k) \). The numbers on the curves are the various dimensionless temperatures \( \bar{T} \). Observing that the values of these curves are almost constant and equal to unity confirms that \( \mu k_B T \) of Eq. \( \text{[11]} \) indeed fulfills almost exactly the SCBA equation \( \text{[20]} \). The inset of Fig. 1 shows \( \kappa_T / \kappa_0 \) as a function of the dimensionless temperature \( \bar{T} \) calculated either by \( \text{[12]} \), corresponding in the figure to the (green) solid and dashed curves, or by the determination of \( \kappa_T / \kappa_0 \) by numerical integration of the right-hand side of \( \text{[13]} \) \( \text{[19]} \). The kink in this curve corresponds to parameter values where \( C_T / \kappa_0 k_{BZ} = 1 \). Note that for graphene and SWNTs we have \( \bar{T}_m \approx 1.34 \) at the melting point \( T_m \approx 5000 \, \text{K} \).

Next we calculate the expectation value \( \langle \nabla_i f \nabla_j f \rangle \) where the average is taken with respect to the Gibbs measure of the Hamiltonian \( \mathcal{H} \) or \( \mathcal{H}^{SCBA} \), respectively. In SCBA, this leads to

\[
\langle \nabla_i f \nabla_j f \rangle_T \approx \frac{1}{(2\pi)^2} \int_{BZ} d^2 k k_i k_j G(k) = \delta_{ij} \bar{T} \kappa_0 k_{BZ}^2 \kappa_0 \ln \left( 1 + \frac{\kappa_0 k_{BZ}^2}{C_T} \right).
\]

(13)

Recalling \( \text{[4]} \) we observe that the strain in the \( xy \)-plane is on the average equal to the negative of \( \text{[13]} \): \( \langle \nabla_i u_i + \nabla_i u_j \rangle_T = -\langle \nabla_i f \nabla_j f \rangle_T \), implying that the self-induced stress due to thermal out-of-plane fluctuations vanishes.

### B. Lindemann numbers

We are not prepared to calculate \((L^2_i) \) of Eq. \( \text{[5]} \) by differentiating the partition function of the elastic Hamiltonian \( \mathcal{H}_i \) twice with respect to the stress source \( \sigma_{ij} \), and setting \( \sigma_{ij} = 0 \) at the end. Going over to the effective Hamiltonian \( \mathcal{H}^{SCBA} \) we obtain two contributions \((L^2_i) \) \( = (L^2_i^{2D}) \) \( = L_i^{2D} \) where the first is the square of the Lindemann number \( \text{[14]} \) for the 2D hexagonal solid given by

\[
(L^2_i^{2D}) = \frac{1}{2} \langle \nabla_i u_m \nabla_i u_m \rangle_{2D} - \langle \nabla_i u_m \rangle_{2D} \langle \nabla_i u_m \rangle_{2D} \approx \frac{k_B T}{\mu} \frac{3 - \nu}{8} k_{BZ}^2.
\]

(14)
FIG. 1: (Color online) Upper figure shows $G^{-1}(k) \approx C_T k^3 + \kappa_i k^4$ divided by the sum of $\kappa_0 k^3$ and the numerical integrated right-hand side of the self-energy function \( \mathcal{S} \) where we used $G^{-1}(k) = C_T k^3 + \kappa_i k^4$. The numbers at the curves are dimensionless temperatures $\tilde{T}$. The inset shows the $\tilde{T}$-behavior of $\kappa_i/\kappa_0$. The low-temperature dashed curve represents Eq. (12) in the regime $C_T \ll \kappa_i k_{BZ}$, the high-temperature solid curve in the regime $C_T \gg \kappa_r k_{BZ}$. The dashed-dotted curve pictures the ratio $\kappa_i/\kappa_0$ obtained from \( \mathcal{S} \) by numerical integration of the right-hand side with $G^{-1}(k) = C_T k^3 + \kappa_i k^4$.

The average $\langle \cdots \rangle_{2D}$ is calculated with respect to the Gibbs measure of the Hamiltonian \( \mathcal{H}_\sigma \) with $f = 0$ corresponding to the 2D crystal. This contribution to $(\mathcal{L}_c^2)^2$ comes from the derivate of $H_{\sigma\sigma}$ in Eq. (16) with respect to $\sigma_{ij}$. The second contribution $\mathcal{L}_c^2$ has its origin in the derivate of the $h_b$ term in (16) with respect to $\sigma_{ij}$, and is found to be

$$L_{c}^{2}(T) \approx \frac{3(1+\nu)^2}{2(2\pi)^2} \int_{BZ} d^2k \left( \frac{k \times q}{q^4} \right)^4 G(k + q) G(k) \Lambda(k + q, k).$$

The vertex correction factor $\Lambda(k + q, k)$ is required within the SCBA by charge-current conservation. We first calculate $L_1$ in the lowest approximation $\Lambda \approx 1$, to be justified below. By using the analytic approximation $L_1$ we obtain for $L_1$ with $L_2$ by integration $L_1$.

$$(17)\\\quad L_{c}^{2}(T) \approx \left\{ \begin{array}{l l} \frac{3(1+\nu)^2}{2(2\pi)^2} T^2 \left( \frac{\kappa_0 k_{BZ}}{\kappa_r} \right)^2 \ln \left( \frac{\kappa_r k_{BZ}}{C_T} \right), & C_T \ll \kappa_r k_{BZ}, \\ 0.3 \frac{3(1+\nu)^2}{32(2\pi)^2} T^2 \left( \frac{\kappa_0 k_{BZ}}{\kappa_r} \right)^2 \left( \frac{\kappa_r k_{BZ}}{C_T} \right)^2, & C_T \gg \kappa_r k_{BZ}. \end{array} \right.$$
modulus $\mu$ will drop to zero, where according to Eqs. \ref{eq:1} and \ref{eq:17}, $L_{1,2}^{1,2D}$ goes to infinity.

Next we calculate the 3D form of \ref{eq:1} ($L_{1,2}^{1,3D} = (L_{1,2}^{1,2D})^2 + L_{2,2}^{2,2D} + \frac{1}{2} \langle \nabla_i f \nabla_j f \rangle_T$) where the last term is due to nearest-neighbor out-of-plane fluctuations given by \ref{eq:13}. The first three terms measure in-plane fluctuations where $L_{2,2}^{2,2D}$ is given by the momentum integral

$$L_{2,2}^{2,2D} = -2(1+\nu) \frac{1}{2(2\pi)^2} \int_{BZ} dq \frac{1}{q^2} \left[ (2\delta_{ij} - 1) q_i q_j - \delta_{ij} \nu q^2 \right] I_{ij}(q) + \frac{1}{2(2\pi)^2} \int_{BZ} dq I_{ij}(q). \tag{18}$$

The functions $I_{ij}(q)$ and $I_{ij,ij}(q)$ denote the Fourier transforms of the expectation values

$$\frac{1}{2} \left[ \langle \nabla_i f(x)f(x)h_f(x') \rangle_T - \langle \nabla_i f(x)\rangle T \langle h_f(x') \rangle T \right]$$

and

$$\frac{1}{4} \left[ \langle \nabla_i f(x)f(x)\nabla_i' f(x') \rangle_T - \langle \nabla_i f(x) \rangle T \langle \nabla_i' f(x') \rangle T \right],$$

respectively. The contribution $L_{2,2}^{2,2D}$ is calculated in the same way as $L_{2,1}^{1,2D}$ \ref{eq:19}, yielding

$$L_{2,2}^{2,2D} \approx \begin{cases} \frac{3}{10(2\pi)^2} \tilde{\beta}^2 \left( \frac{\kappa_0^2 \rho \mu}{\kappa_0^2 C_T^2} \right)^2, & C_T \ll \kappa_0 k_{BZ}, \\ \frac{3(1+\nu)}{10(2\pi)^2} \tilde{\beta}^2 \left( \frac{\kappa_0^2 \rho \mu}{\kappa_0^2 C_T^2} \right)^2, & C_T \gg \kappa_0 k_{BZ}. \end{cases} \tag{19}$$

Here we have used the abbreviation $(1 + \tilde{\beta}) \equiv 1 + (1 + \nu)[-0.31 + 0.25 \nu/(1 + \nu)]$. Using the material parameters of SWNTs and graphene given above, we find that the main contribution to $L_{1,2}^{1,3D}$ comes from the out-of-plane fluctuations and is given by $L_{1,2}^{1,3D} \approx (\langle \nabla_i f \nabla_j f \rangle_T / 2)^{1/2} \approx 0.22$ at the melting point $T_m \approx 5000 K$. Thus, we find that the out-of-plane fluctuations $(\langle \nabla_i f \nabla_j f \rangle_T / 2)^{1/2}$ are even larger than the dominant contribution to the in-plane fluctuations $L_{1,2}^{1,2D}$. By comparing the temperature dependence \ref{eq:13} with \ref{eq:13} we obtain that this is even the case for smaller temperatures. Furthermore, we realize that in contrast to the Lindemann number $L_{1,2}$, the abrupt increase of the Lindemann number $L_{1,2}^{1,3D}$ gives no good signal for the melting point of a solid membrane. The reason is that the vanishing elastic shear modulus $\mu$ at melting contributes in two ways to the dominant fluctuation term $\langle \nabla_i f \nabla_j f \rangle_T / 2$ \ref{eq:13} but neither of them changes this value much at melting. First, the out-of-plane fluctuations depend on $\mu$ via $\kappa_0 / \kappa_0$ and remains finite for $\mu \to 0$, and second they depend pick up logarithmic dependence on $\mu$ from $C_T / \kappa_0 k_{BZ}$.

Consider now the higher-order vertex corrections collected in the factor $\Lambda(k + \mathbf{q}, k)$ in Eq. \ref{eq:16}. First we note that for $\Lambda \equiv 1$ we obtain $2\beta I(q)/k_B T < 3/8$ in the dominant integration regime of \ref{eq:15} near $q \approx C_T / \kappa$ for $C_T \ll \kappa_0 k_{BZ}$ and near $q \approx k_{BZ}$ for $C_T \gg \kappa_0 k_{BZ}$. The factor $3/8$ comes mainly from the reduction of the phase space integral by the projections $P_T$ in the polarisator. We expect that the $n$th order in $\Lambda$ contributes roughly with a factor $[2\beta I(q)/k_B T]^n$ to $I(q)$ in the dominant integration regime of \ref{eq:15} due to the additional phase space projection terms $P_T$. We have checked this explicitly by taking into account first-order corrections in the vertex $\Lambda$ in \ref{eq:16}. A similar suppression of higher-order vertex correction contributions occurs in $I_{ij}(q)$ and $I_{ij,ij}(q)$.

C. Melting temperature

Let us finally discuss the impact of the large out-of-plane fluctuations upon the melting temperature. In Ref. \ref{eq:8} we have calculated the melting temperature of a 2D triangular lattice approximately from the intersection of high- and low-temperature expansion of the free energies associated with the Hamiltonian \ref{eq:13} with zero vertical displacements $f(x)$. The transition was caused by integer-valued defect gauge fields accounting for the plastic deformations of the crystal in the $xy$-plane. These are coupled minimally to the $xy$-displacement fields $u_i(x)$. In that theory, the melting temperature $T_m$ was found to obey the equation

$$\tilde{\beta} \equiv \frac{1}{k_B T_m (2\pi)^2} \approx 0.6, \tag{20}$$

where $v_F \equiv \sqrt{3} a^2 / 2$ denotes the 2D-volume (area) of the fundamental cell. In SWNTs and graphene, this result is modified by a factor $S e^{-2W}$, where $S$ is a structure factor and $e^{-2W}$ a Debye-Waller-like factor caused by the out-of-plane fluctuations $f(x) \neq 0$. The honeycomb lattice of SWNTs contains two atoms per triangular fundamental cell leading to a structure factor $S = 1/2$. To estimate the size of $e^{-2W}$ we observe that in the defect melting model, the defect gauge field appears at a similar place as the vertical distortion $\nabla_i f \nabla_i f / 2$ in the Hamiltonian \ref{eq:13}. Thus one can immediately write down the Hamiltonian $H_p$ of Eq. \ref{eq:8} coming from defects. This leads to the low-temperature expansion of the partition function. In the high-temperature expansion, there exist a dual stress representation of the partition function \ref{eq:7} \ref{eq:9}. In both low- and high-temperature representations, the coupling terms between the defect fields or the stress fields to the out-of-plane fluctuations $f(x)$ are smaller than the pure defect and stress term by approximately a factor $(4\pi^2 \tilde{\beta}^2)^2 L_{2,2}^{2,2D}$ and $(2\pi)^2 L_{2,2}^{2,2D}$, respectively. When neglecting these small coupling terms we find that the partition function receives a sizable correction factor only in the the low-temperature approximation due to the Fock energy of the Hamiltonian $H_p$. The Hartree energy is missing as a consequence of the open-boundary conditions on the membrane \ref{eq:13}. From these considera-
tions we obtain

\[
W = \frac{v_F}{4} \frac{1}{(2\pi)^2} \int dk \Sigma(k) G(k) \]

(21)

\[
= \frac{1}{2} \frac{C_T}{\kappa_r k_{BZ}} \left[ 1 - \frac{C_T}{\kappa_r k_{BZ}} \ln \left( 1 + \frac{\kappa_r k_{BZ}}{C_T} \right) \right] 
+ \frac{1}{2} \left( 1 - \kappa_r \right) \left[ \frac{1}{2} - \frac{C_T}{\kappa_r k_{BZ}} + \left( \frac{C_T}{\kappa_r k_{BZ}} \right)^2 \ln \left( 1 + \frac{\kappa_r k_{BZ}}{C_T} \right) \right].
\]

Using the parameters above for (5,5) SWNTs we obtain \( W \approx 0.06 \) at \( T \approx 5000 \) K. The factor \( e^{-2W} \) gives thus only a small correction to the melting temperature determined by \( W \). The explicit evaluation of that equation yields a melting temperature \( T_m \approx 8000 \) K (\( W \approx 0.075 \)), somewhat larger than the melting temperature \( T_m \approx 5000 \) K of Zhang et al. \([14]\) obtained by numerical simulation.

### III. CONCLUSION

In this paper we have calculated the fluctuations of solid membranes like graphene and single-walled carbon nanotubes with the help of the self-consistent Born-approximation. Our results show that the out-of-plane fluctuations are much larger than the in-plane fluctuations even at low temperatures. Thus they may be expected to have dramatic consequences for the Lindemann numbers as well as the melting temperature of solid membranes in comparison to 2D crystals. Surprisingly, for the melting temperature this expectation was not confirmed. The fluctuations were discussed by evaluating the 3D-version \( \xi^\ast_{1,3D} \) of the Lindemann number \([1]\), originally introduced to estimate the melting temperature of 2D solids, and the Lindemann number \( \xi^\ast_1 \) defined in Eq. \([2]\), originally introduced in cluster physics. We observed that a Lindemann criterion based on \( \xi^\ast_1 \) is more reliable than that based on the former. The associated Lindemann number is dominated by in-plane fluctuations, in contrast to the former which is dominated by the large out-of-plane fluctuations. By calculating, in addition, the melting temperature from a simple defect model of melting for single-walled carbon nanotubes and graphene \([20]\) we observed in contrast to the expectation, that the melting temperature depends only very little on the large out-of-plane fluctuations.

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