Electronic Supplementary Material (ESI) for Nanoscale.

Supplement S1: Derivation of the effective potential acting on stiff monolayers driven over commensurate substrates

SUMMARY

In the following, we shall present a detailed account of our analytical calculation of the total mean force exerted on the center of mass of the colloidal monolayer in the harmonic approximation. For the convenience of those readers that are not anxious to delve into all the details of these derivations, we provide the following quick summary.

For large stiffness of the colloidal layer, i.e., large repulsive Yukawa interaction strength, we regard the build-up phase of a hopping wave as given by a field of small displacement vectors $\mathbf{u}_l$ of the colloidal particles from their ideal lattice sites $\mathbf{R}_l$ on top of a collective rigid translation R of this lattice in the direction x of the external driving force. We argue that during this build-up phase, the monolayer is in quasistatic equilibrium, such that statistical mechanics can be employed to compute the thermally averaged mean force acting on its center of mass. Furthermore, as we assume that the displacements $\mathbf{u}_l$ will be small in relation to the ideal lattice constant of the monolayer, it is justified to employ a harmonic approximation for this task. For periodic boundary conditions, the canonical distribution of the resulting harmonic Hamiltonian factorizes into independent "phonon" contributions, whose energy contributions and covariances are completely determined by the underlying dynamical matrix. It is then straightforward to calculate the averaged covariances σ_{xx}, σ_{xy} and σ_{yy} of the displacement components. To harmonic accuracy, it turns out that these covariances also fully determine the effective total force acting on the center of mass of the monolayer. The final section provides some useful formulas for a practical numerical implementation.

EQUATION OF MOTION OF THE COLLOIDAL MONOLAYER

For overdamped Langevin dynamics, the equations of motion of an N-particle system can be written as

$$\gamma \frac{\mathrm{d}\mathbf{r}_p}{\mathrm{d}t} = \mathbf{F}_p(\vec{\mathbf{r}}) \tag{1}$$

for $p = 0, 1, \ldots, N-1$, where the total force

$$\mathbf{F}_p(\vec{\mathbf{r}}) = \sum_{p' \neq p} \mathbf{F}_{\mathrm{yuk}}(\mathbf{r}_{p'} - \mathbf{r}_p) + \mathbf{F}_{\mathrm{sub}}(\mathbf{r}_p) + \mathbf{F}_d + \mathbf{F}^p_{\mathrm{rand}} \tag{2}$$

acting on particle p is the sum of the Yukawa forces exerted by all other particles, the substrate force, the external homogeneous force and the Langevin random force,

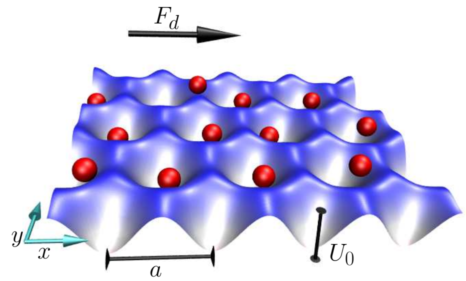

FIG. 1. Schematic view of the potential landscape (12). The direction of the external driving force F_d is indicated by an arrow.

and $\vec{\mathbf{r}}$ is the formal $2N$-dimensional vector built from all position vectors $\mathbf{r}_0, \ldots \mathbf{r}_{N-1}$. The substrate force, $\mathbf{F}_{\mathrm{sub}}(\mathbf{r}_p)$, acting on particle i is the negative gradient of the externally applied potential, $U_{\mathrm{sub}}(\mathbf{r}_p) = -(U_0/9)\{3 + 2[\cos(\mathbf{k}_1\mathbf{r}_p) + \cos(\mathbf{k}_2\mathbf{r}_p) + \cos(\mathbf{k}_3\mathbf{r}_p)]\}$, where the $\mathbf{k}$-vectors are chosen from the set $\mathbf{k}_i/\|\mathbf{k}\| \in \{(\sqrt{3}/2, 1/2), (-\sqrt{3}/2, 1/2), (0,1)\}$ with norm $\|\mathbf{k}\| = 4\pi/a\sqrt{3}$. This choice of $\mathbf{k}$-vectors produces a hexagonal arrangement of potential wells with a lattice constant a and lattice vectors $\mathbf{g} \in \{(a,0), (a/2, \sqrt{3}a/2)\}$. For a schematic view of the system see Fig. 1.

If we perform an average over all N particles,

$$\frac{\gamma}{N} \sum_{p=0}^{N-1} \frac{\mathrm{d}\mathbf{r}_p}{\mathrm{d}t} = \frac{1}{N} \sum_{p=0}^{N-1} \sum_{p' \neq p} \mathbf{F}_{\mathrm{yuk}}(\mathbf{r}_{p'} - \mathbf{r}_p) + \frac{1}{N} \sum_{p=0}^{N-1} \left[\mathbf{F}_{\mathrm{sub}}(\mathbf{r}_p) + \mathbf{F}_d + \mathbf{F}^p_{\mathrm{rand}}\right] \tag{3}$$

the Yukawa forces cancel in a pairwise manner due to Newton's third law, and we are left with a *single* equation of motion

$$\gamma \frac{\mathrm{d}\mathbf{R}}{\mathrm{d}t} = \frac{1}{N} \sum_{p=0}^{N-1} \mathbf{F}_{\mathrm{sub}}(\mathbf{r}_p) + \mathbf{F}_d + \frac{1}{N} \sum_{p=0}^{N-1} \mathbf{F}^p_{\mathrm{rand}} \tag{4}$$

for the center of mass

$$\mathbf{R} = \frac{1}{N} \sum_{p=0}^{N-1} \mathbf{r}_p \tag{5}$$

of the overdamped monolayer, which resembles that of a single overdamped Brownian diffuser in an external potential. The final term in Eqn. (4) is the sum of all of the random forces acting on the system. It is Gaussian distributed and has a variance of $2k_{\mathrm{B}}T/\sqrt{N}$.

Although solving the coupled equations of motion for N interacting particles analytically without any approximation is an impossible task, our claim is that there are exist a series of approximations that simplify Equation 4 sufficiently to obtain accurate theoretical predictions. The motion of the center of mass of the monolayer is governed by the constant driving force, the average substrate force, and the total random Langevin force acting on the monolayer. Although the last two forces are trivial, the first, is not. The difficulty lies in the fact that the substrate force acting on the center of mass when it is located at $\mathbf{R}$, $\bar{\mathbf{F}}(\mathbf{R}) = N^{-1}\sum_{p=0}^{N-1}\mathbf{F}_{\mathrm{sub}}(\mathbf{r}_p,\mathbf{R})$ depends on the positions of every particle in the system, which in turn depend upon their mutual interactions as well as the external forces acting on them. We therefore propose a statistical treatment of the problem. If one imagines a trajectory consisting of an arbitrarily large number of buildup phases, then the mean velocity of the monolayer is,

$$\gamma\left\langle\frac{\mathrm{d}\mathbf{R}}{\mathrm{d}t}\right\rangle = \left\langle\bar{\mathbf{F}}\right\rangle_{\mathbf{R}} + \mathbf{F}_d, \quad (6)$$

where the average, $\langle ...\rangle$, is taken over multiple build up phases. So far, no tangible simplification to the system has been made, other than that the motion of the center of mass of the monolayer during the build up phase can be thought of as the motion of a single particle exposed to an effective substrate force $\mathbf{F}_{\mathrm{eff}}(\mathbf{R}) = \left\langle\bar{\mathbf{F}}\right\rangle_{\mathbf{R}}$. If the distribution of $\bar{\mathbf{F}}$ is narrow, then it can be replaced by its mean value $\mathbf{F}_{\mathrm{eff}}(\mathbf{R})$ in the equation of motion for the center of mass,

$$\gamma\frac{\mathrm{d}\mathbf{R}}{\mathrm{d}t} = \mathbf{F}_{\mathrm{eff}}(\mathbf{R}) + \mathbf{F}_d + \bar{\mathbf{F}}_{\mathrm{random}}, \quad (7)$$

we justify this simplification *a posteriori* in the supplement S2.

QUASISTATIC EQUILIBRIUM

In order to learn something about the functional form of $\mathbf{F}_{\mathrm{eff}}(\mathbf{R})$, we make two assumptions. First, we assume that configurations from different buildup phases with the same $\mathbf{R}$ are not only statistically independent, but are drawn from the equilibrium distribution of the system. We justify this assumption from the fact that the monolayer travels along the substrate walls very slowly. The second assumption we make is that the interparticle potential is so large that the total potential energy of the system can be approximated by a second order Taylor expansion.

The first assumption is that the monolayer, during the buildup phase, moves so slowly that for a given value of $\mathbf{R}$, the probability of observing a microstate obeys a Boltzmann distribution,

$$d\rho(\vec{\mathbf{r}},\mathbf{R}) = d^N\mathbf{r}\,\delta\left(N^{-1}\sum_i \mathbf{r}_i - \mathbf{R}\right)e^{-\beta U_{\mathrm{tot}}(\vec{\mathbf{r}})}, \quad (8)$$

where $\beta = 1/k_{\mathrm{B}}T$. In order to obtain analytical results, the needs to be simplified further. The second assumption, which is the topic of the next section, will serve this purpose.

THE HARMONIC CRYSTAL

In the absence of any external potential, colloidal particles that interact with one another via a screened repulsive Yukawa potential U_{yuk} tend to form a triangular lattice. In the present work, the particle density is chosen precisely in such a way that this lattice is commensurate with the hexagonal structure of the underlying substrate. The total potential energy of the resulting system is

$$U_{\mathrm{tot}} = \frac{1}{2}\sum_{p\neq p'}^{N-1} U_{\mathrm{yuk}}(\,|\mathbf{r}_p - \mathbf{r}_{p'}|\,) + \sum_{p=0}^{N-1} U_{\mathrm{sub}}(\mathbf{r}_p) - F_d\sum_{p=0}^{N-1} r_p^x. \quad (9)$$

In order to calculate the Boltzmann average $F_{\mathrm{eff}}(R) = \mathbf{F}^x_{\mathrm{eff}}(R)$ defined by this potential analytically further approximations must be made. Among the most successful and widely used in solid state physics is the harmonic approximation, which rests on the idea that particles residing in a crystal lattice will mostly perform only small amplitude vibrations around their equilibrium positions, such that a second order Taylor expansion of the potential with respect to the deviations from these equilibrium positions will already capture most of the relevant physics. By imposing periodic boundary conditions (PBCs) and exploiting the resulting translational invariance of the system, the dynamical problem can then be reformulated in terms of certain collective *phonon* variables defined in Fourier space, which are completely decoupled from each other. At least to a good approximation, the whole procedure thus maps the original problem to a non-interacting one, an enormous simplification for dynamical calculations as well as for doing statistical mechanics.

For our present purposes, the instantaneous position $\mathbf{r}_p$ of an individual colloid will be disassembled as follows. Let $\mathbf{R} = (R, 0)$ denote an arbitrary vector, which we will use to describe the global translation of the triangular colloid layer parallel to the direction of the driving force. $\mathbf{R}_p$ denotes the position of the lattice site that the p^{th} particle is assigned to, and $\mathbf{u}_p$ denotes a small residual displacement of the particle with respect to the underlying lattice. Altogether, we then write

$$\mathbf{r}_p = \mathbf{R} + \mathbf{R}_p + \mathbf{u}_p. \quad (10)$$

In terms of this parametrization

$$U_{\rm tot} = \frac{1}{2}\sum_{p'\neq p}^{N-1} U_{\rm yuk}(|\mathbf{R}_p + \mathbf{u}_p - \mathbf{R}_{p'} - \mathbf{u}_{p'}|) + \sum_{p=0}^{N-1} U_{\rm sub}(\mathbf{R}+\mathbf{R}_p+\mathbf{u}_p) - F_d\sum_{p=0}^{N-1} u_p^x - F_d\left(NR + \sum_{p=0}^{N-1} R_p^x\right). \quad (11)$$

Since the substrate potential is periodic in $\mathbf{R}_p$ and the Yukawa potential depends only on the relative distance between two particles, this simplifies to

$$U_{\rm tot} = \frac{1}{2}\sum_{p\neq p'}^{N-1} U_{\rm yuk}(\,|\mathbf{R}_{pp'} + \mathbf{u}_p - \mathbf{u}_{p'}|\,) + \sum_{p=0}^{N-1} U_{\rm sub}(\mathbf{R}+\mathbf{u}_p) - F_d\sum_{p=0}^{N-1} u_p^x - F_d\left(NR + \sum_{p=0}^{N-1} R_p^x\right), \quad (12)$$

where $\mathbf{R}_{pp'} = \mathbf{R}_{p'} - \mathbf{R}_p$ is the difference vector between lattice site p and p'.

In the high coupling limit, when the inter-particle interaction strength is large, the deviations $\mathbf{u}_p$ of the particle positions from the ideal lattice sites $\mathbf{R}_p$ are typically small, so a second order Taylor expansion

$$U_{\rm tot} = \frac{1}{2}\sum_{l,l'=0}^{N-1}\sum_{\mu,\nu} u_l^\mu\,\phi_{\mu\nu}^{ll'}\,u_{l'}^\nu + \frac{\pi}{a}F_{\rm max}\sum_{l=0}^{N-1}\left\{\cos\frac{2\pi R}{a}\left((u_l^x)^2 + \frac{1}{3}(u_l^y)^2\right) + \frac{2}{3}(u_l^y)^2\right\}$$
$$+\left(F_{\rm max}\sin\frac{2\pi R}{a} - F_d\right)\sum_{l=0}^{N-1} u_l^x + C(R,\{\mathbf{R}_p\}), \quad (13)$$

with respect to $\mathbf{u}_p$ may yield a good approximation to the total energy of the system. Unlike the particle induces p and p', the indicies l and l' denote *lattice site* induces that can be equal to each other in the double sum above, as can be seen in the definition of $\phi_{\mu\nu}^{ll'}$,

$$\phi_{\mu\nu}^{ll'} = \frac{\partial^2}{\partial u_l^\mu \partial u_{l'}^\nu}\sum_{p\neq p'}^{N-1} U_{\rm yuk}(\,|\mathbf{u}_p - \mathbf{u}_{p'} + R_{pp'}|\,)|_{\mathbf{u}_p=\mathbf{u}_{p'}=\mathbf{0}}. \quad (14)$$

$C(R,\{\mathbf{R}_p\})$ is the value of the total potential when all $\mathbf{u}_l$ are zero, and $F_{\rm max} = 24\pi k_{\rm B}T/a$ is the maximum force the substrate is able to exert. Using the definition of the substrate potential, the 2×2 matrix of the second derivatives of the external substrate

$$\psi(R) = \frac{2\pi F_{\rm max}}{3a}\begin{pmatrix} 3\cos\frac{2\pi R}{a} & 0 \\ 0 & \cos\frac{2\pi R}{a} + 2\end{pmatrix} 1 \quad (15)$$

turns out to be diagonal when evaluated at $\mathbf{R} = (R,0)$. Collecting linear and quadratic terms, we rewrite (13) in the compact form

$$U_{\rm tot} = \frac{1}{2}\sum_{l,l'=0}^{N-1}\sum_{\mu,\nu} u_l^\mu\,\bar{\mathbb{D}}_{\mu\nu}^{ll'}(R)\,u_{l'}^\nu \quad (16)$$
$$+\left(F_{\rm max}\sin\frac{2\pi R}{a} - F_d\right)\sum_{l=0}^{N-1} u_l^x + C(R,\{\mathbf{R}_l\})$$

where we have set

$$\bar{\mathbb{D}}_{\mu\nu}^{ll'}(R) \equiv \phi_{\mu\nu}^{ll'} + \delta^{ll'}\psi_{\mu\nu}(R). \quad (17)$$

Translational invariance allows to further reduce

$$\frac{1}{2}\sum_{l,l'=0}^{N-1}\sum_{\mu,\nu} u_l^\mu\,\bar{\mathbb{D}}_{\mu\nu}^{ll'}(R)\,u_{l'}^\nu = \frac{N}{2}\sum_{l=0}^{N-1}\sum_{\mu,\nu} u_l^\mu\,\bar{\mathbb{D}}_{\mu\nu}^{l0}(R)\,u_0^\nu \quad (18)$$

Within equilibrium statistical mechanics, our N-particle system is described by the unnormalized probability measure

$$d\rho(\vec{\mathbf{u}}) = d^N\mathbf{u}\,e^{-\beta U_{\rm tot}(\vec{\mathbf{u}})}, \quad (19)$$

where $\beta = 1/k_{\rm B}T$, $\vec{\mathbf{u}}$ is the formal $2N$-dimensional vector built from all displacement vectors $\mathbf{u}_0,\ldots\mathbf{u}_{N-1}$, and $d^N\mathbf{u} = \prod_{l=0}^{N-1} d^2u_l$. Imposing the constraint that the sum of all the u_l^x be zero amounts to considering the restricted probability measure

$$d\rho(\vec{\mathbf{u}}|R) = d^N\mathbf{u}\,\delta\left(\sum_l u_l^x\right)e^{-\beta U_{\rm tot}(\vec{\mathbf{u}})}$$
$$= d^N\mathbf{u}\,\delta\left(\sum_l u_l^x\right)\exp\left\{-\frac{\beta}{2}\vec{\mathbf{u}}^T\tilde{\mathbb{D}}\vec{\mathbf{u}}\right\}. \quad (20)$$

Ensemble averages of observables $A(\{\mathbf{u}_l\})$ are given by

$$\langle A(\vec{\mathbf{u}})\rangle\big|_R = \frac{1}{Z(R)}\int d^N\mathbf{u}\,A(\vec{\mathbf{u}})d\rho(\vec{\mathbf{u}}|R), \quad (21)$$

whose normalization $Z(R) = \int d^N\mathbf{u}\,d\rho(\vec{\mathbf{u}}|R)$ may be called a restricted canonical partition function.

DISCRETE FOURIER TRANSFORM

In contrast to (19), the measure (20) is *not* a simple Gaussian one, as the variables $\mathbf{u}_l$ are not independent due to the delta constraint imposed. However, we now show that by virtue of a discrete Fourier transform, (20) can actually be factorized into simple components, which clears the way for analytical calculations.

As a prerequisite, we introduce the first Brilloun zone $\mathcal{B}$ of the underlying hexagonal lattice with a total number of $N = N_x N_y$ particles. For simplicity, we choose $N_x = N_y = \sqrt{N}$, and furthermore, without loss of generality, we assume N_x and N_y to be even numbers. By definition, $\mathcal{B}$ consists of all cosets represented by wave vectors that are commensurate with the imposed boundary conditions, two such representatives considered as equivalent if they differ by an arbitrary reciprocal vector. For periodic boundary conditions in both the x and y direction, the allowed representatives are

$$q_x = \frac{2\pi}{a}\frac{n_x}{N_x}, \qquad q_y = \frac{4\pi}{\sqrt{3}a}\frac{n_y}{N_y}, \tag{22}$$

where n_x and n_y are integers. A convenient choice of a set of representative wave vectors for $\mathcal{B}$ is provided by the Voronoi cell around a point in reciprocal space (see Fig. 2).

Now we can introduce the discrete Fourier transform

$$u_l^\mu = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}\in\mathcal{B}} \tilde{u}^\mu(\mathbf{q}) e^{i\mathbf{q}\mathbf{R}_l}. \tag{23}$$

We have

$$\sum_{l=0}^{N-1} u_l^x = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}\in\mathcal{B}} \tilde{u}^x(\mathbf{q}) \underbrace{\sum_l e^{i\mathbf{q}\mathbf{R}_l}}_{N\delta(\mathbf{q},\mathbf{0})} = \sqrt{N}\tilde{u}^x(\mathbf{0}). \tag{24}$$

Furthermore

$$\begin{aligned} &\frac{1}{2}\sum_{l,l'=0}^{N-1}\sum_{\mu,\nu} u_l^\mu\, \bar{\mathbb{D}}_{\mu\nu}^{ll'}(R)\, u_{l'}^\nu \\ &= \frac{1}{2N}\sum_{l,l'=0}^{N-1}\sum_{\mathbf{q},\mathbf{q}'}\sum_{\mu,\nu} \tilde{u}^\mu(\mathbf{q}) e^{i\mathbf{q}\mathbf{R}_l} \bar{\mathbb{D}}_{\mu\nu}^{ll'}(R) \tilde{u}^\nu(\mathbf{q}') e^{i\mathbf{q}'\mathbf{R}_l'} \\ &= \frac{1}{2}\sum_{\mathbf{q},\mathbf{q}'}\sum_{\mu,\nu} \tilde{u}^\mu(\mathbf{q}) \underbrace{\left[\frac{1}{N}\sum_{l,l'=0}^{N-1} e^{i\mathbf{q}\mathbf{R}_l}\bar{\mathbb{D}}_{\mu\nu}^{ll'}(R) e^{i\mathbf{q}'\mathbf{R}_l'}\right]}_{\equiv \bar{\mathbb{D}}_{\mu\nu}(\mathbf{q},\mathbf{q}',R)} \tilde{u}^\nu(\mathbf{q}'). \end{aligned} \tag{25}$$

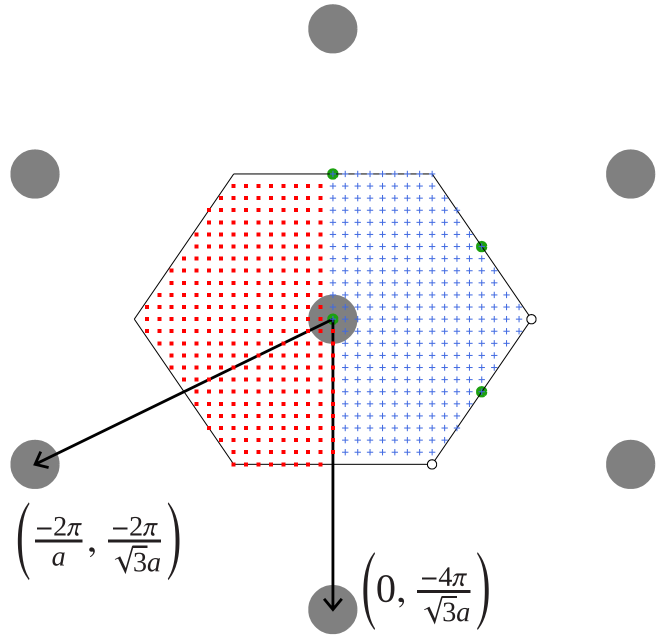

FIG. 2. $\mathbf{q}$-vectors (points) for a 24×24 hexagonal monolayer. The gray dots are lattice sites belonging to the reciprocal lattice of the original lattice. The four green circles mark the special points defined in Eqn. (37) and belong to $\mathcal{B}_0$. The blue crosses belong to $\mathcal{B}_+$ and the red boxes belong to the set $\mathcal{B}_-$. The empty circles at two of the vertices of the small hexagon are points that must be omitted from to avoid double counting, since they differ from already included vertex points by a reciprocal lattice vector.

Using translational invariance, we write

$$\begin{aligned} \bar{\mathbb{D}}_{\mu\nu}(\mathbf{q},\mathbf{q}',R) &= \frac{1}{N}\sum_{l,l'=0}^{N-1} e^{i\mathbf{q}\mathbf{R}_l}\bar{\mathbb{D}}_{\mu\nu}^{l-l',0}(R) e^{i\mathbf{q}'\mathbf{R}_l'} \\ &= \frac{1}{N}\sum_{l,l'=0}^{N-1} e^{i\mathbf{q}(\mathbf{R}_l+\mathbf{R}_l')}\bar{\mathbb{D}}_{\mu\nu}^{l,0}(R) e^{i\mathbf{q}'\mathbf{R}_l'} \\ &= \frac{1}{N}\sum_{l=0}^{N-1} e^{i\mathbf{q}\mathbf{R}_l}\bar{\mathbb{D}}_{\mu\nu}^{l,0}(R) \underbrace{\sum_{l'=0}^{N-1} e^{i(\mathbf{q}+\mathbf{q}')\mathbf{R}_l'}}_{=N\delta_{\mathbf{q}+\mathbf{q}',\mathbf{0}}} \end{aligned} \tag{26}$$

This suggests we define

$$\bar{\mathbb{D}}_{\mu\nu}(\mathbf{q},R) := \sum_{l=0}^{N-1} \bar{\mathbb{D}}_{\mu\nu}^{l,0}(R) e^{i\mathbf{q}\mathbf{R}_l}. \tag{27}$$

Additional symmetry of $\bar{\mathbb{D}}_{\mu\nu}^{ll'}(R)$ under exchange of $l \leftrightarrow l'$ also yields $\bar{\mathbb{D}}_{\mu\nu}^{l,0}(R) = \bar{\mathbb{D}}_{\mu\nu}^{0,l}(R) = \bar{\mathbb{D}}_{\mu\nu}^{-l,0}(R)$, and thus

$$\left[\bar{\mathbb{D}}_{\mu\nu}(\mathbf{q},R)\right]^* = \sum_{l=0}^{N-1} \bar{\mathbb{D}}_{\mu\nu}^{-l,0}(R) e^{-i\mathbf{q}\mathbf{R}_l} = \bar{\mathbb{D}}_{\mu\nu}(\pm\mathbf{q},R), \tag{28}$$

i.e., $\bar{\mathbb{D}}_{\mu\nu}(\mathbf{q}, R) \in \mathbb{R}$, a fact that could also be anticipated from the manifest reality of

$$\frac{1}{2}\sum_{l=0,l'}^{N-1}\sum_{\mu,\nu} u_l^\mu\, \bar{\mathbb{D}}_{\mu\nu}^{ll'}(R)\, u_{l'}^\nu \tag{29}$$

$$= \frac{1}{2}\sum_{\mathbf{q},\mathbf{q}'\in\mathcal{B}}\sum_{\mu,\nu} \tilde{u}^\mu(\mathbf{q})\bar{\mathbb{D}}_{\mu\nu}(\mathbf{q}', R)\delta_{\mathbf{q}+\mathbf{q}',\mathbf{0}}\tilde{u}^\nu(\mathbf{q}')$$

$$= \frac{1}{2}\sum_{\mathbf{q}\in\mathcal{B}}\sum_{\mu,\nu} \tilde{u}^\mu(-\mathbf{q})\bar{\mathbb{D}}_{\mu\nu}(\mathbf{q}, R)\tilde{u}^\nu(\mathbf{q}). \tag{30}$$

From the definition of the discrete Fourier transform (23) it follows immediately that since the components u_l^μ are real-valued, their Fourier amplitudes must obey

$$\tilde{u}^\mu(-\mathbf{q}) = [\tilde{u}^\mu(\mathbf{q})]^*. \tag{31}$$

Thus, if we introduce the complex two-dimensional vector

$$\tilde{\mathbf{u}}(\mathbf{q}) = \begin{pmatrix} \tilde{u}^x(\mathbf{q}) \\ \tilde{u}^y(\mathbf{q}) \end{pmatrix} \tag{32}$$

and its adjoint

$$\tilde{\mathbf{u}}^+(\mathbf{q}) = \begin{pmatrix} \tilde{u}^x(-\mathbf{q}) & \tilde{u}^y(-\mathbf{q}) \end{pmatrix}, \tag{33}$$

we obtain the compact formula

$$\frac{1}{2}\sum_{l=1,l'}^{N}\sum_{\mu,\nu} u_l^\mu\, \bar{\mathbb{D}}_{\mu\nu}^{ll'}(R)\, u_{l'}^\nu = \frac{1}{2}\sum_{\mathbf{q}\in\mathcal{B}} \tilde{\mathbf{u}}^+(\mathbf{q}) \cdot \bar{\mathbb{D}}(\mathbf{q}, R) \cdot \tilde{\mathbf{u}}(\mathbf{q}). \tag{34}$$

The delta function constraint in the measure (20) is rewritten as

$$\delta\left(\tilde{u}^x(\mathbf{0})\right) = \delta\left(\sqrt{N}\tilde{u}^x(\mathbf{0})\right) = \frac{\delta\left(\tilde{u}^x(\mathbf{0})\right)}{\sqrt{N}}. \tag{35}$$

Finally, we need to express the volume element $d^N\mathbf{u}$ appearing in the measure (20) in terms of the Fourier amplitudes (23). Taking the real part of (23), we obtain

$$u_l^\mu = \frac{1}{\sqrt{N}}\sum_{\mathbf{q}\in\mathcal{B}} [\Re\tilde{u}^\mu(\mathbf{q}) + i\Im\tilde{u}^\mu(\mathbf{q})][\cos(\mathbf{q}\mathbf{R}_l) + i\sin(\mathbf{q}\mathbf{R}_l)]$$

$$= \sum_{\mathbf{q}} \left[\frac{\cos(\mathbf{q}\mathbf{R}_l)}{\sqrt{N}}\Re\tilde{u}^\mu(\mathbf{q}) - \frac{\sin(\mathbf{q}\mathbf{R}_l)}{\sqrt{N}}\Im\tilde{u}^\mu(\mathbf{q})\right], \tag{36}$$

which is reminiscent of an orthogonal transformation, except that we seem to have doubled the number of variables. To avoid such a double-counting, note that the reality condition $\tilde{u}^\mu(-\mathbf{q}) = [\tilde{u}^\mu(\mathbf{q})]^*$ implies that $\Re\tilde{u}^\mu(-\mathbf{q}) = \Re\tilde{u}^\mu(\mathbf{q})$ and $\Im\tilde{u}^\mu(-\mathbf{q}) = -\Re\tilde{u}^\mu(\mathbf{q})$, are linearly dependent. The latter condition requires care. By definition, two vectors of the first Brilloun zone are regarded as equal if they differ by a reciprocal lattice vector $\mathbf{G}$. But this implies the possibility that $\mathbf{q}$ and $-\mathbf{q}$ can well be representatives of the same coset of $\mathcal{B}$. A trivial case is the zero vector $\mathbf{Q}_0 = \mathbf{0}$, but in our hexagonal lattice this applies to three more so-called "special high-symmetry vectors". We shall denote the subset of $\mathcal{B}$ that holds these four vectors

$$\mathbf{Q}_0 = (0, 0), \qquad \mathbf{Q}_1 = \left(0, \frac{2\pi}{\sqrt{3}a}\right),$$
$$\mathbf{Q}_2 = \left(\frac{\pi}{a}, \frac{\pi}{\sqrt{3}a}\right), \qquad \mathbf{Q}_3 = \left(\frac{\pi}{a}, -\frac{\pi}{\sqrt{3}a}\right), \tag{37}$$

as $\mathcal{B}_0$. Of course, for $\mathbf{Q} \in \mathcal{B}_0$ the Fourier amplitudes $\tilde{\mathbf{u}}(\mathbf{Q})$ must be real as can be directly understood from observing that $\mathbf{Q} \cdot \mathbf{R}_l$ is an integer multiple of π. The residual $N - 4$ elements of the Brilloun zone are given by distinct pairs of representatives $(\mathbf{q}, -\mathbf{q})$ and can now be organized into two subsets $\mathcal{B}_\pm$ of positive and negative "parity" by any convenient definition. This leads to a partition

$$\mathcal{B} = \mathcal{B}_0 \cup \mathcal{B}_+ \cup \mathcal{B}_- \tag{38}$$

of the total Brilloun zone. In Fig. 2, we have illustrated such a partition of $\mathcal{B}$ into subsets with zero (green circled points), positive (blue points) and negative (red points) parity. In this notation, (36) may be rewritten in a more concise way as

$$u_l^\mu = \sum_{\pi(\mathbf{Q})\in\mathcal{B}_0} \frac{(\pm 1)}{\sqrt{N}}\tilde{u}^\mu(\mathbf{Q}) \tag{39}$$
$$+ \sum_{\mathbf{q}\in\mathcal{B}_+} \left[\frac{2\cos(\mathbf{q}\mathbf{R}_l)}{\sqrt{N}}\Re\tilde{u}^\mu(\mathbf{q}) - \frac{2\sin(\mathbf{q}\mathbf{R}_l)}{\sqrt{N}}\Im\tilde{u}^\mu(\mathbf{q})\right].$$

As this construction reveals, a new set of $2N = 2[4 + 2 \times (N-4)/2]$ independent real variables is given by

$$\tilde{u}^\mu(\mathbf{Q}), \quad \mathbf{Q} \in \mathcal{B}_0, \quad \mu = x, y \tag{40}$$
$$\Re\tilde{u}^\mu(\mathbf{q}),\ \Im\tilde{u}^\mu(\mathbf{q}), \quad \mathbf{q} \in \mathcal{B}_+, \quad \mu = x, y. \tag{41}$$

In terms of these variables, we can rewrite

$$\frac{1}{2}\sum_{\mathbf{q}\in\mathcal{B}} \tilde{\mathbf{u}}^+(\mathbf{q}) \cdot \bar{\mathbb{D}}(\mathbf{q}, R) \cdot \tilde{\mathbf{u}}(\mathbf{q}) = \frac{1}{2}\sum_{\mathbf{Q}\in\mathcal{B}_0} \tilde{u}^\mu(\mathbf{Q})\bar{\mathbb{D}}_{\mu\nu}(\mathbf{Q}, R)\tilde{u}^\nu(\mathbf{Q}) + \sum_{\mathbf{q}\in\mathcal{B}_+} [\tilde{u}^\mu(\mathbf{q})]^*\bar{\mathbb{D}}_{\mu\nu}(\mathbf{q}, R)u^\nu(\mathbf{q})$$
$$= \frac{1}{2}\sum_{\mathbf{Q}\in\mathcal{B}_0} \tilde{u}^\mu(\mathbf{Q})\bar{\mathbb{D}}_{\mu\nu}(\mathbf{Q}, R)\tilde{u}^\nu(\mathbf{Q}) + \sum_{\mathbf{q}\in\mathcal{B}_+} [\Re\tilde{u}^\mu(\mathbf{q}) - i[\Im\tilde{u}^\mu(\mathbf{q})]^*]\bar{\mathbb{D}}_{\mu\nu}(\mathbf{q}, R)[\Re u^\nu(\mathbf{q}) + i\Im u^\nu(\mathbf{q})]. \tag{42}$$

In the last line the imaginary contributions must cancel identically for the sum to be real, and we obtain

$$\begin{aligned}
&\frac{1}{2}\sum_{\mathbf{q}\in\mathcal{B}} \tilde{\mathbf{u}}^{+}(\mathbf{q})\cdot\bar{\mathbb{D}}(\mathbf{q},R)\cdot\tilde{\mathbf{u}}(\mathbf{q}) && (43)\\
&=\frac{1}{2}\sum_{\mathbf{Q}\in\mathcal{B}_0}\bar{\mathbb{D}}_{\mu\nu}(\mathbf{Q},R)\tilde{u}^{\mu}(\mathbf{Q})\tilde{u}^{\nu}(\mathbf{Q})+\sum_{\mathbf{q}\in\mathcal{B}_+}\bar{\mathbb{D}}_{\mu\nu}(\mathbf{q},R)[\Re\tilde{u}^{\mu}(\mathbf{q})\Re u^{\nu}(\mathbf{q})+\Im\tilde{u}^{\mu}(\mathbf{q})\Im u^{\nu}(\mathbf{q})] \\
&=\frac{1}{2}\sum_{\mathbf{Q}\in\mathcal{B}_0}\tilde{\mathbf{u}}^{T}(\mathbf{Q})\bar{\mathbb{D}}(\mathbf{Q},R)\tilde{\mathbf{u}}(\mathbf{Q})+\sum_{\mathbf{q}\in\mathcal{B}_+}\Re\tilde{\mathbf{u}}^{T}(\mathbf{q})\bar{\mathbb{D}}(\mathbf{q},R)\Re\tilde{\mathbf{u}}(\mathbf{q})+\sum_{\mathbf{q}\in\mathcal{B}_+}\Im\tilde{\mathbf{u}}^{T}(\mathbf{q})\bar{\mathbb{D}}(\mathbf{q},R)\Im\tilde{\mathbf{u}}(\mathbf{q}). && (44)
\end{aligned}$$

As to the volume element in the measure (20), we have

$$d^N\mathbf{u}=J\cdot\prod_{\mathbf{Q}\in\mathcal{B}_0}d^2\tilde{u}(\mathbf{Q})\cdot\prod_{\mathbf{q}\in\mathcal{B}_+}d^2\Re\tilde{u}(\mathbf{Q})d^2\Im\tilde{u}(\mathbf{Q}) \tag{45}$$

where J is the determinant of the Jacobi matrix of the discrete Fourier transformation, and an additional factor arises from the delta function constraint, since

$$\delta\left(\tilde{u}^x(\mathbf{0})\right)=\delta\left(\sqrt{N}\tilde{u}^x(\mathbf{0})\right)=\frac{\delta\left(\tilde{u}^x(\mathbf{0})\right)}{\sqrt{N}}. \tag{46}$$

Writing down the unnormalized probability measure

$$d\tilde{\rho}(\{\tilde{\mathbf{u}}(\mathbf{q})\}|R)=\prod_{\mathbf{Q}\in\mathcal{B}_0}d^2\tilde{u}(\mathbf{Q})\cdot\prod_{\mathbf{q}\in\mathcal{B}_+}d^2\Re\tilde{u}(\mathbf{q})d^2\Im\tilde{u}(\mathbf{q})\cdot\delta\left(\tilde{u}^x(\mathbf{0})\right)e^{-\frac{\beta}{2}\sum_{\mathbf{q}\in\mathcal{B}}\tilde{\mathbf{u}}^{+}(\mathbf{q})\cdot\bar{\mathbb{D}}(\mathbf{q},R)\cdot\tilde{\mathbf{u}}(\mathbf{q})} \tag{47}$$

which obviously factorizes into

$$d\tilde{\rho}(\{\tilde{\mathbf{u}}(\mathbf{q})\}|R)=\prod_{\mathbf{Q}\in\mathcal{B}_0}d\tilde{\rho}^{(0)}(\tilde{\mathbf{u}}(\mathbf{Q})|R)\cdot\prod_{\mathbf{q}\in\mathcal{B}_+}d\tilde{\rho}^{(R)}(\tilde{\mathbf{u}}(\mathbf{q})|R)\cdot d\tilde{\rho}^{(I)}(\tilde{\mathbf{u}}(\mathbf{q})|R) \tag{48}$$

as promised, where

$$\begin{aligned}
d\tilde{\rho}(\tilde{\mathbf{u}}(\mathbf{0})|R)&=d^2\tilde{u}(\mathbf{0})\delta\left(\tilde{u}^x(\mathbf{0})\right)e^{-\frac{\beta}{2}\tilde{\mathbf{u}}^{T}(\mathbf{0})\bar{\mathbb{D}}(\mathbf{0},R)\tilde{\mathbf{u}}(\mathbf{0})}, && (49)\\
d\tilde{\rho}^{(0)}(\tilde{\mathbf{u}}(\mathbf{Q})|R)&=d^2\tilde{u}(\mathbf{Q})e^{-\frac{\beta}{2}\tilde{\mathbf{u}}^{T}(\mathbf{Q})\bar{\mathbb{D}}(\mathbf{Q},R)\tilde{\mathbf{u}}(\mathbf{Q})}, \qquad \mathbf{Q}\in\mathcal{B}_0 && (50)\\
d\tilde{\rho}^{(R)}(\tilde{\mathbf{u}}(\mathbf{q})|R)&=d^2\Re\tilde{u}(\mathbf{q})e^{-\beta\Re\tilde{\mathbf{u}}^{T}(\mathbf{q})\bar{\mathbb{D}}(\mathbf{q},R)\Re\tilde{\mathbf{u}}(\mathbf{q})}, \qquad \mathbf{q}\in\mathcal{B}_+ && (51)\\
d\tilde{\rho}^{(I)}(\tilde{\mathbf{u}}(\mathbf{q})|R)&=d^2\Im\tilde{u}(\mathbf{q})e^{-\beta\Im\tilde{\mathbf{u}}^{T}(\mathbf{q})\bar{\mathbb{D}}(\mathbf{q},R)\Im\tilde{\mathbf{u}}(\mathbf{q})}, \qquad \mathbf{q}\in\mathcal{B}_+. && (52)
\end{aligned}$$

EXPLICIT FORMULAE

Having fully characterized the coordinate transformation that transforms the dynamical matrix of the harmonic crystal into Fourier space, we now present the explicit form of $\bar{\mathbb{D}}(\mathbf{q})$. To this end, we make use of the abbreviations,

$$\begin{aligned}
C_x(\mathbf{q})&=\cos\left(\frac{a}{2}q_x\right),\qquad S_x(\mathbf{q})=\sin\left(\frac{a}{2}q_x\right), && (53)\\
C_y(\mathbf{q})&=\cos\left(\frac{a\sqrt{3}}{2}q_y\right),\ S_y(\mathbf{q})=\sin\left(\frac{a\sqrt{3}}{2}q_y\right), && (54)\\
f&=\frac{2\pi F_{\max}}{3a},\qquad g=\frac{1}{r}U'_{\text{yuk}}(r)|_{r=a}, && (55)\\
h&=\left[U''_{\text{yuk}}(r)-\frac{1}{r}U'_{\text{yuk}}(r)\right]_{r=a}. && (56)
\end{aligned}$$

The quantities are the result of performing a Fourier transformation on a hexagonal lattice and the quantities in Eqn. (56) are the coupling parameters of the particles in the monolayer. These parameters depend upon the first and second derivatives of the substrate potential and the inter-particle interactions. For the Yukawa potential,

$$g = -\frac{\tilde{\Gamma} e^{-\kappa a}}{a^3}[1+\kappa a] = -\frac{\Gamma}{a^2}(1+\kappa a), \tag{57}$$

$$h = \frac{\tilde{\Gamma} e^{-\kappa a}}{a^3}[3+3\kappa a+(\kappa a)^2] = \frac{\Gamma}{a^2}[3+3\kappa a+(\kappa a)^2]. \tag{58}$$

As in previous work, the coupling strength between two colloids, Γ, is the potential energy between two particles that are separated by one lattice constant. Thus $\Gamma := \tilde{\Gamma}\frac{e^{-\kappa a}}{a}$. We note that for all allowed values of Γ, a, and κ, the absolute value of g is strictly larger than that of h. Furthermore, since these expressions are not quadratic in a, we do not express them in terms of the density of the system, $\rho^{-1} = N^{-1}\frac{\sqrt{3}}{2}a^2$, although that might be a more natural definition.

In terms of this parametrization, we find that

$$\bar{\mathbb{D}}(\mathbf{q}) = \begin{pmatrix} D_0(\mathbf{q},R)+D_{11}(\mathbf{q},R) & D_{12}(\mathbf{q},R) \\ D_{12}(\mathbf{q},R) & D_0(\mathbf{q},R) \end{pmatrix} \tag{59}$$

where

$$D_0(\mathbf{q},R) = 4g\left[2 - C_x^2(\mathbf{q}) - C_x(\mathbf{q})C_y(\mathbf{q})\right] + 3h\left[1 - C_x(\mathbf{q})C_y(\mathbf{q})\right] + f\left[2+\cos(2\pi R/a)\right] \tag{60}$$

$$D_{11}(\mathbf{q},R) = 2h\left[1 + C_x(\mathbf{q})C_y(\mathbf{q}) - 2C_x^2(\mathbf{q})\right] - 4f\sin^2(\pi R/a) \tag{61}$$

$$D_{12}(\mathbf{q},R) = \sqrt{3}\, h S_x(\mathbf{q})S_y(\mathbf{q}). \tag{62}$$

The results we shall derive below will involve the elements of the inverse matrix

$$\bar{\mathbb{G}}(\mathbf{q},R) := [\beta\bar{\mathbb{D}}(\mathbf{q},R)]^{-1} = \frac{1}{\beta \det \bar{\mathbb{D}}(\mathbf{q})}\begin{pmatrix} D_0(\mathbf{q}) & -D_{12}(\mathbf{q}) \\ -D_{12}(\mathbf{q}) & D_0(\mathbf{q})+D_{11}(\mathbf{q}) \end{pmatrix}. \tag{63}$$

Calculation of covariances

We now look at the expectation values

$$\sigma_{\mu\nu}(R) = \left\langle \frac{1}{N}\sum_l u_l^\mu u_l^\nu \right\rangle \Big|_R. \tag{64}$$

Since

$$\frac{1}{N}\sum_{l=0}^{N-1} u_l^\mu u_l^\nu = \frac{1}{N^2}\sum_{\mathbf{q},\mathbf{q}'} \tilde{u}^\mu(\mathbf{q})\tilde{u}^\nu(\mathbf{q}') \underbrace{\sum_l e^{i(\mathbf{q}+\mathbf{q}')\mathbf{R}_l}}_{N\delta(\mathbf{q}+\mathbf{q}',\mathbf{0})} = \sum_{\mathbf{q}} \tilde{u}^\mu(\mathbf{q})\tilde{u}^\nu(-\mathbf{q}), \tag{65}$$

this reduces to

$$\begin{aligned}
\sigma_{\mu\nu}(R) &= \frac{1}{N}\sum_{\mathbf{q}} \langle \tilde{u}^\mu(\mathbf{q})\tilde{u}^\nu(-\mathbf{q})\rangle \big|_R \\
&= \frac{1}{N}\sum_{\mathbf{Q}\in\mathcal{B}_0} \langle \tilde{u}^\mu(\mathbf{Q})\tilde{u}^\nu(\mathbf{Q})\rangle \big|_R + \frac{1}{N}\sum_{\mathbf{q}\in\mathcal{B}_+} \langle \tilde{u}^\mu(\mathbf{q})\tilde{u}^\nu(-\mathbf{q})\rangle \big|_R + \frac{1}{N}\sum_{\mathbf{q}\in\mathcal{B}_-} \langle \tilde{u}^\mu(\mathbf{q})\tilde{u}^\nu(-\mathbf{q})\rangle \big|_R \\
&= \frac{1}{N}\sum_{\mathbf{Q}\in\mathcal{B}_0} \langle \tilde{u}^\mu(\mathbf{Q})\tilde{u}^\nu(\mathbf{Q})\rangle \big|_R + \frac{2}{N}\sum_{\mathbf{q}\in\mathcal{B}_+} \Re\langle \tilde{u}^\mu(\mathbf{q})\tilde{u}^\nu(-\mathbf{q})\rangle \big|_R \\
&= \frac{1}{N}\sum_{\mathbf{Q}\in\mathcal{B}_0} \langle \tilde{u}^\mu(\mathbf{Q})\tilde{u}^\nu(\mathbf{Q})\rangle \big|_R + \frac{2}{N}\sum_{\mathbf{q}\in\mathcal{B}_+} \langle \Re\tilde{u}^\mu(\mathbf{q})\Re\tilde{u}^\nu(\mathbf{q}) + \Im\tilde{u}^\mu(\mathbf{q})\Im\tilde{u}^\nu(\mathbf{q})\rangle \big|_R.
\end{aligned} \tag{66}$$

To compute these expectation values, we use the well-known formula

$$\frac{\int d^D x\, x_i x_j\, e^{-\frac{1}{2}\mathbf{x}^T\mathbf{A}\mathbf{x}}}{\int d^D x\, e^{-\frac{1}{2}\mathbf{x}^T\mathbf{A}\mathbf{x}}} = A^{-1}_{ij} \tag{67}$$

valid for Gaussian integrals, which yields

$$\langle \tilde{u}^\mu(\mathbf{Q})\tilde{u}^\nu(\mathbf{Q})\rangle\big|_R = \bar{\mathbb{G}}^{\mu\nu}(\mathbf{Q},R), \qquad \mathbf{Q}\in\mathcal{B}_0 \tag{68}$$

$$\langle \Re\tilde{u}^\mu(\mathbf{q})\Re\tilde{u}^\nu(\mathbf{q})\rangle\big|_R = \langle \Im\tilde{u}^\mu(\mathbf{q})\Im\tilde{u}^\nu(\mathbf{q})\rangle\big|_R = \frac{\bar{\mathbb{G}}^{\mu\nu}(\mathbf{q},R)}{2}, \qquad \mathbf{q}\in\mathcal{B}_+ \tag{69}$$

Due to the presence of the delta constraint, special care has to be taken for $\mathbf{Q} = \mathbf{0}$. Since the matrix $\bar{\mathbb{D}}(\mathbf{q},R)$ is actually diagonal for $\mathbf{q} = \mathbf{0}$ (cf. Eqn. (62) below), we have

$$\langle \tilde{u}^\mu(\mathbf{0})\tilde{u}^\nu(\mathbf{0})\rangle\big|_R = \begin{cases} 1/\beta\bar{\mathbb{D}}^{yy}(\mathbf{0},R), & \mu=\nu=y \\ 0, & \text{else}\end{cases} = \begin{cases} \bar{\mathbb{G}}^{yy}(\mathbf{0},R), & \mu=\nu=y \\ 0, & \text{else}\end{cases} \tag{70}$$

In summary, we have shown that

$$\begin{aligned}\sigma_{yy}(R) &= \frac{1}{N}\sum_{\mathbf{Q}\in\mathcal{B}_0}\bar{\mathbb{G}}^{yy}(\mathbf{Q},R) + \frac{2}{N}\sum_{\mathbf{q}\in\mathcal{B}_+}\frac{\bar{\mathbb{G}}^{yy}(\mathbf{q},R)}{2} + \frac{2}{N}\sum_{\mathbf{q}\in\mathcal{B}_+}\frac{\bar{\mathbb{G}}^{yy}(\mathbf{q},R)}{2} \\ &= \frac{1}{N}\sum_{\mathbf{Q}\in\mathcal{B}_0}\bar{\mathbb{G}}^{yy}(\mathbf{Q},R) + \frac{2}{N}\sum_{\mathbf{q}\in\mathcal{B}_+}\bar{\mathbb{G}}^{yy}(\mathbf{q},R) \\ &= \frac{1}{N}\sum_{\mathbf{Q}\in\mathcal{B}_0}\bar{\mathbb{G}}^{yy}(\mathbf{Q},R) + \frac{1}{N}\sum_{\mathbf{q}\in\mathcal{B}_+}\bar{\mathbb{G}}^{yy}(\mathbf{q},R) + \sum_{\mathbf{q}\in\mathcal{B}_-}\bar{\mathbb{G}}^{yy}(\mathbf{q},R)\end{aligned} \tag{71}$$

i.e.,

$$\sigma_{yy}(R) = \frac{1}{N}\sum_{\mathbf{q}\in\mathcal{B}}\bar{\mathbb{G}}^{yy}(\mathbf{q},R). \tag{72}$$

In the same way we can show that

$$\sigma_{xx}(R) = \frac{1}{N}\sum_{\mathbf{q}\in\mathcal{B}\setminus\{\mathbf{0}\}}\bar{\mathbb{G}}^{xx}(\mathbf{q},R). \tag{73}$$

and also

$$\sigma_{xy}(R) = \frac{1}{N}\sum_{\mathbf{q}\in\mathcal{B}\setminus\{\mathbf{0}\}}\bar{\mathbb{G}}^{xy}(\mathbf{q},R). \tag{74}$$

However, a closer examination reveals that due to the special structure of the matrix elements

$$\bar{\mathbb{G}}^{xy}(\mathbf{q},R) = -\frac{\sqrt{3}hS_x(\mathbf{q})S_y(\mathbf{q})}{\beta\det\bar{\mathbb{D}}(\mathbf{q})} \tag{75}$$

the sum above actually vanishes. To show this, we first observe that the contributions of all $\mathbf{q}$-vectors of the types $(q_x,0),(0,q_y)$ and $(q_x,2\pi/a\sqrt{3})$ are zero because of the vanishing product of the sine functions $S_x(\mathbf{q})S_y(\mathbf{q})$. The remaining contributions from the wavevectors $\mathbf{q}\in\mathcal{B}_+\cup\mathcal{B}_-$ can be organized into pairs of $(q_x,q_y),(q_x,-q_y)$, whose contributions mutually cancel (note that the numerator of (75) assumes different signs for the vectors in each couple, whereas the sign of the determinant in the denominator remains the same). In retrospect, the fact that the cross correlation $\sigma_{xy}(R)$ is found to be zero within the harmonic approximation could have been anticipated from the fact that the dynamical matrix is constructed from (i) the sum of a pairwise central potential and (ii) a substrate potential with vanishing mixed second derivatives along the path $(R,0)$.

In summary we have the covariances

$$\sigma_{xx}(R) = \frac{1}{N}\sum_{\mathbf{q}\in\mathcal{B}\setminus\{\mathbf{0}\}}\bar{\mathbb{G}}^{xx}(\mathbf{q},R) \tag{76}$$

$$\sigma_{yy}(R) = \frac{1}{N}\sum_{\mathbf{q}\in\mathcal{B}}\bar{\mathbb{G}}^{yy}(\mathbf{q},R) \tag{77}$$

$$\sigma_{xy}(R) = 0 \tag{78}$$

which are the results announced in Eqn. (8) of the main paper.

MEAN FORCE

By symmetry, the only nonzero component of the total force acting on the center of mass of the monolayer (7) located at $\mathbf{R} = (R,0)$ is along the x-direction. Before averaging, this component of is

$$F_{\text{eff}}(R) = \frac{F_{\max}}{N}\sum_{l=0}^{N-1}\sin[k_x(u_l^x+R)]\cos(k_y u_l^y). \tag{79}$$

Here $k_x = 2\pi/a$ and $k_y = 2\pi/a\sqrt{3}$. Using the trigonometric identity $\sin(x+y) = \sin x \cos y + \cos x \sin y$, we rewrite this as

$$
\begin{aligned}
F_{\rm eff}(R) &= \frac{F_{\max}}{N}\cos(k_x R)\sum_{l=0}^{N-1}\sin(k_x u_l^x)\cos(k_y u_l^y) + \frac{F_{\max}}{N}\sin(k_x R)\sum_{l=0}^{N-1}\cos(k_x u_l^x)\cos(k_y u_l^y) \\
&= \frac{F_{\max}}{N}\cos(k_x R)\sum_{l=0}^{N-1}\frac{e^{ik_x u_l^x} - e^{-ik_x u_l^x}}{2i}\frac{e^{ik_y u_l^y} + e^{-ik_y u_l^y}}{2} \\
&\quad + \frac{F_{\max}}{N}\sin(k_x R)\sum_{l=0}^{N-1}\frac{e^{ik_x u_l^x} + e^{-ik_x u_l^x}}{2}\frac{e^{ik_y u_l^y} + e^{-ik_y u_l^y}}{2} \qquad (80) \\
&= \frac{F_{\max}\cos(k_x R)}{4i}\frac{1}{N}\sum_{l=0}^{N-1}\left[e^{i(k_x u_l^x + k_y u_l^y)} + e^{i(k_x u_l^x - k_y u_l^y)} - e^{i(-k_x u_l^x + k_y u_l^y)} - e^{-i(k_x u_l^x + k_y u_l^y)}\right] \\
&\quad + \frac{F_{\max}\sin(k_x R)}{4}\frac{1}{N}\sum_{l=0}^{N-1}\left[e^{i(k_x u_l^x + k_y u_l^y)} + e^{i(k_x u_l^x - k_y u_l^y)} + e^{i(-k_x u_l^x + k_y u_l^y)} + e^{-i(k_x u_l^x + k_y u_l^y)}\right].
\end{aligned}
$$

If we define the four wave vectors

$$
\mathbf{k}_{(\pm\pm)} := \begin{pmatrix} \pm k_x \\ \pm k_y \end{pmatrix}, \qquad (81)
$$

we can restate this as

$$
\begin{aligned}
F_{\rm eff}(R) &= \frac{F_{\max}\cos(k_x R)}{4i}\frac{1}{N}\sum_{l=0}^{N-1}\left[e^{i\mathbf{k}_{(++)}\mathbf{u}_l} + e^{i\mathbf{k}_{(+-)}\mathbf{u}_l} - e^{i\mathbf{k}_{(-+)}\mathbf{u}_l} - e^{i\mathbf{k}_{(--)}\mathbf{u}_l}\right] \\
&\quad + \frac{F_{\max}\sin(k_x R)}{4}\frac{1}{N}\sum_{l=0}^{N-1}\left[e^{i\mathbf{k}_{(++)}\mathbf{u}_l} + e^{i\mathbf{k}_{(+-)}\mathbf{u}_l} + e^{i\mathbf{k}_{(-+)}\mathbf{u}_l} + e^{i\mathbf{k}_{(--)}\mathbf{u}_l}\right]. \qquad (82)
\end{aligned}
$$

To compute the $F_{\rm eff}(R)$ of the mean net force, we work out the averages

$$
\langle e^{i\mathbf{k}_{(\pm\pm)}\mathbf{u}_l}\rangle|_R = \frac{\int d^N\mathbf{u}\,\delta\left(\sum u_m^x\right)\exp\left\{-\frac{\beta}{2}\vec{\mathbf{u}}^T\tilde{\mathbb{D}}\vec{\mathbf{u}} + i\mathbf{u}_l^T\cdot\mathbf{k}_{(\pm\pm)}\right\}}{\int d^N\mathbf{u}\,\delta\left(\sum u_m^x\right)\exp\left\{-\frac{\beta}{2}\vec{\mathbf{u}}^T\tilde{\mathbb{D}}\vec{\mathbf{u}}\right\}}. \qquad (83)
$$

At this point it would be straightforward to follow the prescription of the preceding paragraph, i.e., we could rewrite the above integral in terms of the real variables $\Re\tilde{\mathbf{u}}(\mathbf{q}), \Im\tilde{\mathbf{u}}(\mathbf{q})$ and utilize the well-known Gaussian formula

$$
\frac{\int d^D x\, e^{-\frac{1}{2}\mathbf{x}^T\mathbf{A}\mathbf{x} + \boldsymbol{b}^T\cdot\mathbf{x}}}{\int d^D x\, e^{-\frac{1}{2}\mathbf{x}^T\mathbf{A}\mathbf{x}}} = e^{\frac{1}{2}\sum_{ij} b_i A_{ij}^{-1} b_j}. \qquad (84)
$$

An alternative evaluation proceeds by completion of squares in (83). First, slightly symmetrize this expression, rewriting it as

$$
\langle e^{i\mathbf{k}_{(\pm\pm)}\mathbf{u}_l}\rangle|_R = \frac{\int d^N\mathbf{u}\,\delta\left(\sum u_m^x\right)\exp\left\{-\frac{\beta}{2}\vec{\mathbf{u}}^T\tilde{\mathbb{D}}\vec{\mathbf{u}} + \frac{i}{2}\mathbf{u}_l^T\cdot\mathbf{k}_{(\pm\pm)} + \frac{i}{2}\mathbf{k}_{(\pm\pm)}^T\cdot\mathbf{u}_l\right\}}{\int d^N\mathbf{u}\,\delta\left(\sum u_m^x\right)\exp\left\{-\frac{\beta}{2}\vec{\mathbf{u}}^T\tilde{\mathbb{D}}\vec{\mathbf{u}}\right\}}. \qquad (85)
$$

With the help of (23) and (34) this is restated as

$$
\begin{aligned}
& -\frac{\beta}{2}\vec{\mathbf{u}}^T\beta\tilde{\mathbb{D}}\vec{\mathbf{u}} + \frac{i}{2}\mathbf{u}_l^T\mathbf{k}_{(\pm\pm)} + \frac{i}{2}\mathbf{k}_{(\pm\pm)}\mathbf{u}_l \qquad (86) \\
&= \sum_{\mathbf{q}}\left[\tilde{\mathbf{u}}^+(\mathbf{q})\cdot\frac{\beta\bar{\mathbb{D}}(\mathbf{q},R)}{2}\cdot\tilde{\mathbf{u}}(\mathbf{q}) + \frac{i}{2}\tilde{\mathbf{u}}^+(\mathbf{q})\frac{\mathbf{k}_{(\pm\pm)}e^{-i\mathbf{q}\mathbf{R}_l}}{\sqrt{N}} + \frac{i}{2}\frac{\mathbf{k}_{(\pm\pm)}e^{i\mathbf{q}\mathbf{R}_l}}{\sqrt{N}}\tilde{\mathbf{u}}(\mathbf{q})\right] \\
&= -\frac{1}{2}\sum_{\mathbf{q}}\left[\tilde{\mathbf{u}}^+(\mathbf{q})\cdot\beta\bar{\mathbb{D}}(\mathbf{q},R)\cdot\tilde{\mathbf{u}}(\mathbf{q}) - i\tilde{\mathbf{u}}^+(\mathbf{q})\cdot\bar{\mathbb{E}}(\mathbf{q};l) - i\bar{\mathbb{E}}^+(\mathbf{q};l)\cdot\tilde{\mathbf{u}}(\mathbf{q})\right],
\end{aligned}
$$

where we have introduced the abbreviation

$$\bar{\mathbb{E}}(\mathbf{q};l) := \frac{\mathbf{k}_{(\pm\pm)}e^{-i\mathbf{q}\mathbf{R}_l}}{\sqrt{N}}. \tag{87}$$

For all nonzero $\mathbf{q}$ we now apply the identity

$$\boldsymbol{U}^+\boldsymbol{D}\boldsymbol{U} + \boldsymbol{U}^+\boldsymbol{V} + \boldsymbol{V}^+\boldsymbol{U} = (\boldsymbol{U}^+ + \boldsymbol{V}^+\boldsymbol{D}^{-1})\boldsymbol{D}(\boldsymbol{U} + \boldsymbol{D}^{-1}\boldsymbol{V}) - \boldsymbol{V}^+\boldsymbol{D}^{-1}\boldsymbol{V} \tag{88}$$

in the form

$$\begin{aligned}
&\tilde{\mathbf{u}}^+(\mathbf{q})\cdot\beta\bar{\mathbb{D}}(\mathbf{q},R)\cdot\tilde{\mathbf{u}}(\mathbf{q}) - i\bar{\mathbb{E}}^+(\mathbf{q};l)\cdot\tilde{\mathbf{u}}(\mathbf{q}) - i\tilde{\mathbf{u}}^+(\mathbf{q})\cdot\bar{\mathbb{E}}(\mathbf{q};l)\\
&= \left[\tilde{\mathbf{u}}^+(\mathbf{q}) - i\bar{\mathbb{E}}^+(\mathbf{q};l)\bar{\mathbb{G}}(\mathbf{q},R)\right]\cdot\beta\bar{\mathbb{D}}(\mathbf{q},R)\cdot\left[\tilde{\mathbf{u}}(\mathbf{q}) - i\bar{\mathbb{G}}(\mathbf{q},R)\bar{\mathbb{E}}(\mathbf{q};l)\right] + \bar{\mathbb{E}}^+(\mathbf{q};l)\cdot\bar{\mathbb{G}}(\mathbf{q},R)\cdot\bar{\mathbb{E}}(\mathbf{q};l)\\
&= \left[\tilde{\mathbf{u}}^+(\mathbf{q}) - i\bar{\mathbb{E}}^+(\mathbf{q};l)\bar{\mathbb{G}}(\mathbf{q},R)\right]\cdot\beta\bar{\mathbb{D}}(\mathbf{q},R)\cdot\left[\tilde{\mathbf{u}}(\mathbf{q}) - i\bar{\mathbb{G}}(\mathbf{q},R)\bar{\mathbb{E}}(\mathbf{q};l)\right] + \frac{1}{N}\mathbf{k}^T_{(\pm\pm)}\cdot\bar{\mathbb{G}}(\mathbf{q},R)\cdot\mathbf{k}_{(\pm\pm)}.
\end{aligned} \tag{89}$$

Note that the l-dependence has disappeared from the last contribution. On the other hand, due to the delta function constraint, the $\mathbf{q}=\mathbf{0}$ contribution to the above sum

$$\tilde{\mathbf{u}}^+(\mathbf{0})\cdot\beta\bar{\mathbb{D}}(\mathbf{0},R)\cdot\tilde{\mathbf{u}}(\mathbf{0}) - i\tilde{\mathbf{u}}^+(\mathbf{0})\cdot\bar{\mathbb{E}}(\mathbf{0};l) - i\bar{\mathbb{E}}^+(\mathbf{0};l)\cdot\tilde{\mathbf{u}}(\mathbf{0}) \tag{90}$$

actually reduces to

$$\beta\bar{\mathbb{D}}^{yy}(\mathbf{0},R)(\tilde{u}^y(\mathbf{0}))^2 - 2i\tilde{u}^y(\mathbf{0})\frac{k^y_{(\pm\pm)}}{\sqrt{N}}. \tag{91}$$

Since $1/\beta\bar{\mathbb{D}}^{yy}(\mathbf{0},R) = \bar{\mathbb{G}}^{yy}(\mathbf{0},R)0$ (cf. Eqn. (70)), we complete the squares as

$$\beta\bar{\mathbb{D}}^{yy}(\mathbf{0},R)(\tilde{u}^y(\mathbf{0}))^2 - 2i\tilde{u}^y(\mathbf{0})\frac{k^y_{(\pm\pm)}}{\sqrt{N}} = \beta\bar{\mathbb{D}}^{yy}(\mathbf{0},R)\left(\tilde{u}^y(\mathbf{0}) - i\frac{\bar{\mathbb{G}}^{yy}(\mathbf{0},R)k^y_{(\pm\pm)}}{\sqrt{N}}\right)^2 + \frac{1}{N}\bar{\mathbb{G}}^{yy}(\mathbf{0},R)\left(k^y_{(\pm\pm)}\right)^2, \tag{92}$$

which is also independent of l. As anticipated from (84), we end up with

$$\langle e^{i\mathbf{k}_{(\pm\pm)}\mathbf{u}_l}\rangle|_R = \exp\left\{-\frac{1}{2}\mathbf{k}^T_{(\pm\pm)}\cdot\frac{1}{N}\sum_{\mathbf{q}\neq\mathbf{0}}\bar{\mathbb{G}}(\mathbf{q},R)\cdot\mathbf{k}_{(\pm\pm)}\right\}. \tag{93}$$

Comparison with (72)-(73) reveals that this can be rewritten in the compact form

$$\langle e^{i\mathbf{k}_{(\pm\pm)}\mathbf{u}_l}\rangle|_R = \exp\left\{-\mathbf{k}^T_{(\pm\pm)}\cdot\frac{\bar{\sigma}(R)}{2}\cdot\mathbf{k}_{(\pm\pm)}\right\}, \tag{94}$$

where

$$\bar{\sigma}(R) = \begin{pmatrix}\sigma_{xx}(R) & \sigma_{xy}(R)\\ \sigma_{xy}(R) & \sigma_{yy}(R)\end{pmatrix} \tag{95}$$

is the 2×2 matrix of covariances.

In applying these results to (82), the term $\propto\cos(k_xR)$ is identically zero as could have been anticipated from symmetry arguments, since the contributions from the four different vectors $\mathbf{k}_{(\pm\pm)}$ cancel each other. In the second term $\propto\sin(k_xR)$, it is also clear that the terms for $\mathbf{k}_{(--)}$ and $\mathbf{k}_{(-+)}$ will give the same result as those for for $\mathbf{k}_{(--)}$ and $\mathbf{k}_{(+-)}$, respectively. Thus we are left with

$$\begin{aligned}
\langle F_{\rm net}\rangle|_R &= F_d + \frac{F_{\max}\sin(k_xR)}{2}\left(e^{-\mathbf{k}^T_{(++)}\cdot\frac{\bar{\sigma}(R)}{2}\cdot\mathbf{k}_{(++)}} + e^{-\mathbf{k}^T_{(+-)}\cdot\frac{\bar{\sigma}(R)}{2}\cdot\mathbf{k}_{(+-)}}\right)\\
&= F_d + \frac{F_{\max}\sin(k_xR)}{2}\left(e^{-\frac{1}{2}\sigma_{xx}k_x^2-\sigma_{xy}k_xk_y-\frac{1}{2}\sigma_{yy}k_y^2} + e^{-\frac{1}{2}\sigma_{xx}k_x^2+\sigma_{xy}k_xk_y-\frac{1}{2}\sigma_{yy}k_y^2}\right),
\end{aligned} \tag{96}$$

i.e.,

$$F_{\rm eff}(R) = -F_{\max}\sin(k_xR)\cosh\left(\sigma_{xy}k_xk_y\right)\exp\left\{-\frac{1}{2}\left(\sigma_{xx}k_x^2 + \sigma_{yy}k_y^2\right)\right\}. \tag{97}$$

Reverting to the former definitions $k_x = 2\pi/a$ and $k_y = 2\pi/a\sqrt{3}$, we end up with

$$F_{\text{eff}}(R) = -F_{\max} \sin\left(\frac{2\pi a}{R}\right) \cosh\left(\frac{4\pi^2}{\sqrt{3}a^2}\sigma_{xy}\right) \exp\left\{-\frac{2\pi^2}{a^2}\left[\sigma_{xx} + \frac{\sigma_{yy}}{3}\right]\right\}. \tag{98}$$

Taking advantage of the fact that σ_{xy} is zero, we finally obtain,

$$F_{\text{eff}}(R) = -F_{\max} \sin\left(\frac{2\pi a}{R}\right) \exp\left\{-\frac{2\pi^2}{a^2}\left[\sigma_x^2 + \frac{\sigma_y^2}{3}\right]\right\} \tag{99}$$

as presented in Equation 7 of the main paper.