UNIVERSITÀ DEGLI STUDI DI MILANO
FACOLTÀ DI SCIENZE E TECNOLOGIE

Corso di Laurea Magistrale in Fisica

Weak-coupling frictional sliding with phononic dissipation in a semi-infinite crystal

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Anno Accademico 2024-2025

"I do not know what I may appear to the world, but to myself I seem to have been only like a boy playing on the seashore, and diverting myself in now and then finding a smoother pebble or a prettier shell than ordinary, whilst the great ocean of truth lay all undiscovered before me."

Isaac Newton

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16 April 2026

Abstract

We address the problem of dynamical friction by means of analytic many-body techniques. Our model consists of a particle (the "slider") moving on a surface of a 3D semi-infinite crystal interacting weakly with its atoms and therefore exciting phonons. By means of linear response theory, we obtain an explicit expression for the friction force slowing down as a function of its speed.

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Chapter 1

Introduction

Understanding frictional phenomena is a fascinating fundamental problem with huge potential impact on energy saving.

Aristotle was the first to speculate about friction, but only with Leonardo da Vinci and Galileo Galilei, in the centuries XVI and XVII, the first physical experiments was done. They found that friction is a property of contacting objects in relative motion, and not a property of the objects themselves. Over the centuries, at macroscopic scales, friction is been thoroughly studied, and it is known that, when observing the interaction between two surfaces in contact two types of friction can arise: static and dynamic friction. But at atomic scales, analytic friction predictions still a hard problem.

Nanofriction investigates the complex process of transformation of mechanical energy into thermal energy, through excitation of the vibrational degrees of freedom (phonons) of solids, as well as electronic ones, when available. Understanding friction at atomic scale is becoming especially relevant due to the massive development of nanotechnology. The miniaturization of electro-mechanic devices leads to an increasing surface/volume ratio, and the consequent increase to the relative importance of friction. But also the recent experimental interest in the investigation of phononic dynamics in sliding friction [1, 2] through advanced techniques, such as the atomic-force-microscope (AFM), underlines the need of a microscopic theory of phonon coupling and dissipation. Molecular-dynamics (MD) simulations, as long as they are based on accurate force fields, describe well the experimental results [3, 4]. The downside of MD simulations is that they are computationally expensive and they provide a friction evaluation for one (or a few) set of physical parameters, that include at least: the sliding velocity, applied load, and temperature. Whenever any of these physical parameters is modified, a brand new simulation is required. This is a very good reason why it would be desirable to be able to predict friction analytically, so that the whole velocity/load/temperature dependence can be calculated at once.

This thesis addresses the problem of nanofriction in this context. We attack the problem of deriving an analytical expression for the friction of a classical particle (the "slider") that moves on a surface of a 3D semi-infinite crystal interacting weakly with its atoms, e.g. through Van-der-Waals forces, and thus exciting phonons. The slider represents, e.g., the apex atom of a sharp AFM tip, interacting with the surface of an harmonic crystal. We obtain a formula for the average frictional force that slows the slider down, evaluated by means of its loss of energy per unit time, as a function of its velocity and distance from the surface, and of the substrate temperature.

Our work is organized as follows. In Chapter 2 we summarize the state of art of the problem, i.e. the theory of channeling through an infinite crystal. In Chapter 3 we introduce a surface, thus formalizing our model, and we setup its solution based on the linear-response theory (LRT). In Chapter 4 we derive an analytical expression for the friction force based on the retarded density-density response function. Finally, in Chapter 5 we discuss our results and illustrate future extensions of the present research.

Chapter 2

The state of art

In this chapter we summarize the recent advances in weak-coupling frictional sliding problems. Early evaluation of dissipation based on LRT was carried out in the context of non-contact friction, mainly by various research teams led by A.I. Volokitin [5, 6, 7, 8]. Analytic understanding of dynamical friction and dissipation in contact sliding at the atomic scale started with a minimal 1D model [9, 10, 11] and was later extended to 2D and 3D channeling problems [12, 13].

2.1 Sliding along a linear harmonic chain

A first analytical description of the friction force and its dependence on the slider velocity was pioneered in a minimal 1D model. The problem analyzed was introduced and described in Ref. [9]. As sketched in Fig. 2.1, the model consists of a slider, implemented in its simplest form as a pointlike particle characterized by mass M, position x_{SL}, and velocity v_{SL}, interacting weakly via a two-body potential with each atom in a harmonic chain characterized

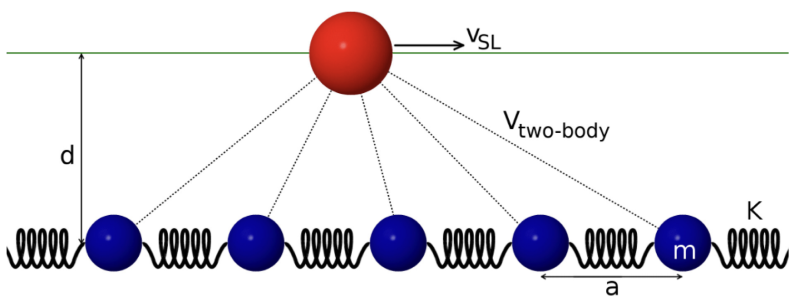

FIGURE 2.1: A sketch of the 1D model. The large sphere represents the slider, which moves along a fixed line (solid line) and interacts via two-body potential with all atoms in the harmonic chain (smaller spheres).

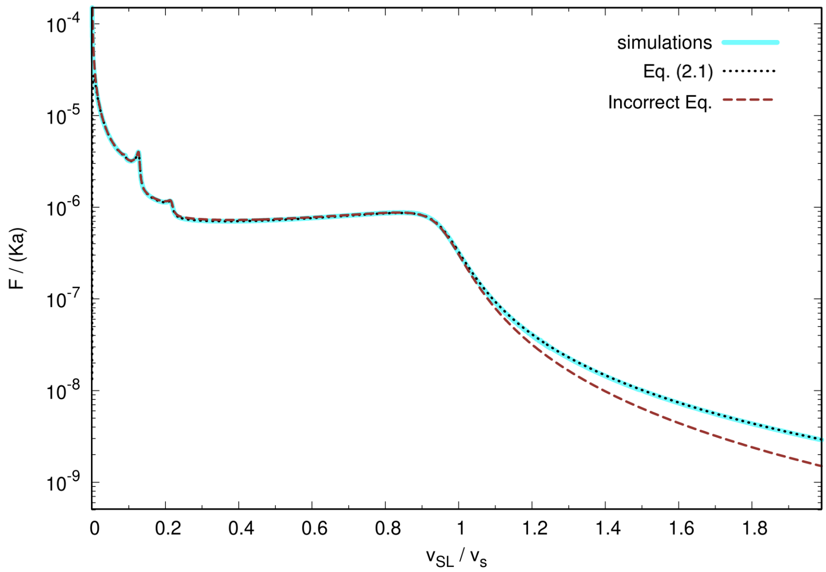

FIGURE 2.2: Comparison of the slider-speed dependence of the friction force $F(v_{\rm SL})$ obtained from the two analytical expressions, the correct Eq. (2.1) (from Ref. [11]) and the incorrect equation of the original paper [10], with that obtained through numerical MD simulations in the same conditions. Exact Eq. (2.1) maintains an excellent agreement at all speeds. Adapted from Ref. [11].

by particles of mass m, nearest-neighbor couplings with spring constant K, and equilibrium spacing a. The slider and the chain atoms move in one dimension (1D) along parallel lines at a fixed distance d. The slider velocity is approximated to a constant value.

A formula for the friction force was obtained by means of analytic linear response theory (LRT):

$$F = \frac{1}{2ma}\int_{-\infty}^{+\infty} dQ \frac{Q^3}{\omega(Q)} |\tilde{V}(Q)|^2 \frac{\frac{\gamma}{2\pi}}{(Q\, v_{\rm SL} - \omega(Q))^2 + \left(\frac{\gamma}{2}\right)^2}\,, \tag{2.1}$$

where $\omega(Q)$ is the dispersion of the sound modes of the harmonic chain, $\tilde{V}(Q)$ is the Fourier transform of the slider-solid interaction potential, and γ is a small frictional damping constant which affects the motion of each chain particle. Note that the integral over Q implies an extended-Brillouin-zone (BZ) scheme for the phonons. For full theory details, see Refs. [10, 11].

The explicit formula (2.1) reproduces the friction measured by classical atomistic simulations very accurately, as shown in Fig. 2.2.

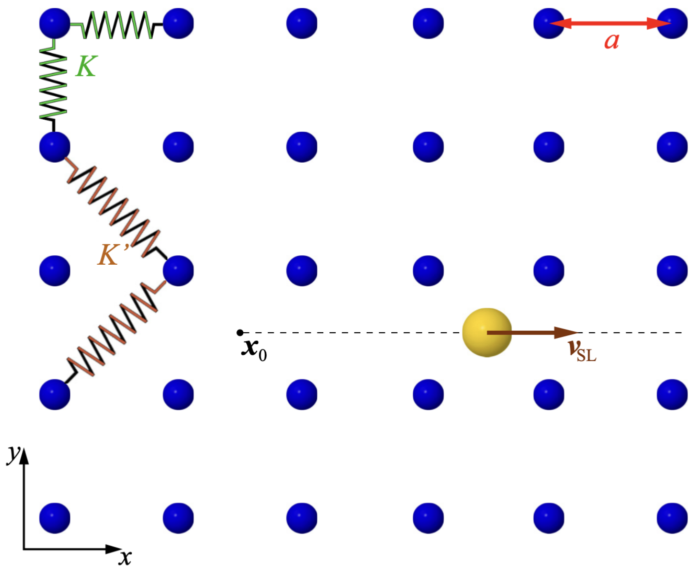

FIGURE 2.3: A sketch of the 2D or 3D model: the slider (large yellow sphere) follows a channeled trajectory (dashed line) inside a crystal. The slider interacts with each atom in the harmonic crystal through a two-body potential $V(r)$. The vibrational properties of the simple-cubic (3D) or square (2D) lattice crystal are determined by first- and second-neighbor springs with elastic constants K and K', respectively. From Ref. [12].

2.2 Channeling through a cubic crystal in 2 or 3 dimensions

The problem of phonon dissipation was recently extended to 2D and 3D crystals, focusing on channeling models [12]. Again, friction is evaluated as the loss of energy per unit time of a channeling particle that interacts weakly, through conservative short-range forces, with a vibrating crystal. A sliding classical particle, traversing the crystal, generates phonons excitations which are described at a quantum level.

As sketched in Fig. 2.3, the model consists of a slider, implemented in its simplest form as a pointlike particle characterized by mass M, position $\mathbf{x}_{SL}$, and velocity $\mathbf{v}_{SL}$ (approximately constant) that follows a channeled trajectory inside the crystal. The slider interacts with each atom in the harmonic system through a two-body potential. The crystal is characterized by particles of mass m that at equilibrium are arranged as a square (2D) or simple-cubic (3D) lattice. Harmonic nearest- and second-neighbor springs with elastic constants K and K' and equilibrium lengths a and $\sqrt{2}a$, respectively, guarantee the mechanical stability and determine the phonon dispersions.

The potential mediates the energy transfer to the harmonic crystal. It needs to be weak to make the perturbative approach meaningful. In a quantum crystal the atomic positions are delocalized in space. As a consequence, the potential energy function $V(r)$ is evaluated at arbitrary values of its argument r:

$$V(r) = V(|\mathbf{x} - \mathbf{x}_{\rm SL}(t)|) = V(|\mathbf{x} - \mathbf{x}_0 - \mathbf{v}_{\rm SL}t|) \equiv V_{ext}(\mathbf{x}, t)\,, \tag{2.2}$$

with $\mathbf{x}_0$ the slider starting point. Therefore, a weak-coupling theory would certainly fail if the interaction diverges rapidly for $r \to 0$. For these channeling problems, it is important to adopt a potential that is Fourier Transform integrable. There are many different potentials that one can adopt, such as a regularized Lennard-Jones (LJ), a Gaussian, or a Woods-Saxon function. In Ref. [12] a regularized LJ potential is adopted.

The average friction force F is evaluated by means of the time-average of the dissipated power by the slider

$$\bar{W} = F\, v_{\rm SL}\,, \tag{2.3}$$

executed over a period $\tau = a/v_{\rm SL}$, which is the time that the slider takes to advance by an interatomic equilibrium length a. LRT allows us to calculate the instantaneous power transferred by the slider to the crystal [14]:

$$W = \frac{d}{dt}E(t) \simeq -\int d^3x \int d^3x' \int_{-\infty}^{+\infty} dt'\, V_{ext}(\mathbf{x}, t)\, \frac{\partial \chi^R_{nn}(\mathbf{x}, \mathbf{x}'; t - t')}{\partial t} \times V_{ext}(\mathbf{x}', t')\,. \tag{2.4}$$

$E(t)$ is the internal energy of the crystal at time t, $\chi^R_{nn}(\mathbf{x}, \mathbf{x}'; t - t')$ is the retarded density-density response function of the unperturbed crystal defined as

$$\chi^R_{nn}(\mathbf{x}, \mathbf{x}'; t - t') = -\frac{i}{\hbar}\theta(t - t')\, \langle [\hat{n}(\mathbf{x}, t - t'), \hat{n}(\mathbf{x}', 0)] \rangle\,, \tag{2.5}$$

and $V_{ext}(\mathbf{x}, t)$ is the perturbation produced on the crystal by the slider at time t. Appendix A summarizes the general formalism of LRT.

Taking advantage of the lattice translation invariance, the external potential and the retarded response function are written in a convenient Fourier representation. The detailed calculation is published in Ref. [12]. Here we report the main analytic result for the friction force. This frictional force is written as a function of the slider speed $v_{\rm SL}$ and the Fourier transform of the slider-crystal-atom interaction potential $\tilde{V}(\mathbf{Q})$:

$$\begin{aligned} F &= \frac{\bar{W}}{v_{\rm SL}} = \frac{1}{\tau}\int_0^\tau dt\, \frac{W(t)}{v_{\rm SL}} \\ &= \frac{4\pi}{ma^3}\sum_{\mathbf{G}_\perp} e^{-i\mathbf{x}_0\cdot\mathbf{G}_\perp}\int \frac{d^3Q}{(2\pi)^3} Q_x \tilde{V}(|\mathbf{Q}|)\tilde{V}(|\mathbf{Q}+\mathbf{G}_\perp|)\times \\ &\quad e^{-W(\mathbf{Q})-W(\mathbf{Q}+\mathbf{G}_\perp)}\sum_\lambda \mathbf{Q}\cdot\boldsymbol{\epsilon}_\lambda(\mathbf{Q})(\mathbf{Q}+\mathbf{G}_\perp)\cdot\boldsymbol{\epsilon}_\lambda(\mathbf{Q})\,\mathcal{L}(\mathbf{Q},v_{\rm SL},\gamma)\,, \end{aligned} \tag{2.6}$$

where

$$\mathcal{L}(\mathbf{Q},v_{\rm SL},\gamma) = \frac{\gamma}{2\pi}\frac{4Q_x v_{\rm SL}}{[(Q_x v_{\rm SL}-\omega_\lambda(\mathbf{Q}))^2+(\frac{\gamma}{2})^2][(Q_x v_{\rm SL}+\omega_\lambda(\mathbf{Q}))^2+(\frac{\gamma}{2})^2]}\,. \tag{2.7}$$

Here, $\omega_\lambda(\mathbf{Q})$ and $\boldsymbol{\epsilon}_\lambda(\mathbf{Q})$ are the crystal's phonon frequencies and polarization vectors, respectively; $\mathbf{G}_\perp$ are the reciprocal-lattice vectors perpendicular to the direction $\hat{\mathbf{x}}$ of the slider velocity; $\mathbf{x}_0$ is the slider's initial position; $e^{-W(\mathbf{Q})}$ are Debye-Waller factors; and γ is a coefficient quantifying the (weak) dissipation leading to phonon decay in the crystal.

A similar result is also obtained in the 2D model:

$$\begin{aligned} F &= \frac{2\pi}{ma^2}\sum_{\mathbf{G}_\perp} e^{-i\mathbf{x}_0\cdot\mathbf{G}_\perp}\int \frac{d^2Q}{(2\pi)^2}\, Q_x\, \tilde{V}(|\mathbf{Q}|)\tilde{V}(|\mathbf{Q}+\mathbf{G}_\perp|)\times \\ &\quad e^{-W(\mathbf{Q})-W(\mathbf{Q}+\mathbf{G}_\perp)}\sum_\lambda \mathbf{Q}\cdot\boldsymbol{\epsilon}_\lambda(\mathbf{Q})(\mathbf{Q}+\mathbf{G}_\perp)\cdot\boldsymbol{\epsilon}_\lambda(\mathbf{Q})\,\mathcal{L}(\mathbf{Q},v_{\rm SL},\gamma)\,. \end{aligned} \tag{2.8}$$

The predictions of Eq. (2.6) and Eq. (2.8) are validated by the striking quantitative agreement with direct energy-loss MD simulations (see Figs. 2.4 and 2.5). In these comparison, zero temperature is considered, both in the MD simulations done to evaluate the friction force and in the analytic expressions (2.6) and (2.8), as obtained by setting the Debye-Waller factors equal to unity, as appropriate in the limit of very low temperature.

Even more recently, N. Gialnisio, in his thesis [13], investigated these Debye-Waller factor as a function of $\mathbf{Q}$ and temperature in the 3D channeling model, see Fig. 2.6. Using these results, he calculated friction taking the appropriate Debye-Waller factors into account for a few temperatures, see Fig. 2.7. No comparison with finite-temperature simulations is available yet.

All these remarkable results take advantage of the full lattice invariance of an infinitely-extended crystal. As a consequence, they do not describe friction between two solids in contact, where certainly one expects nontrivial effects associated to sliding on a surface. Our work aims to investigate precisely this more practically relevant condition: an analytic description for the friction experienced by a sliding object, such as an atomic-force-microscope (AFM) tip, gently grazing a flat crystal surface. This is the problem that we proceed to address in the next Chapter.

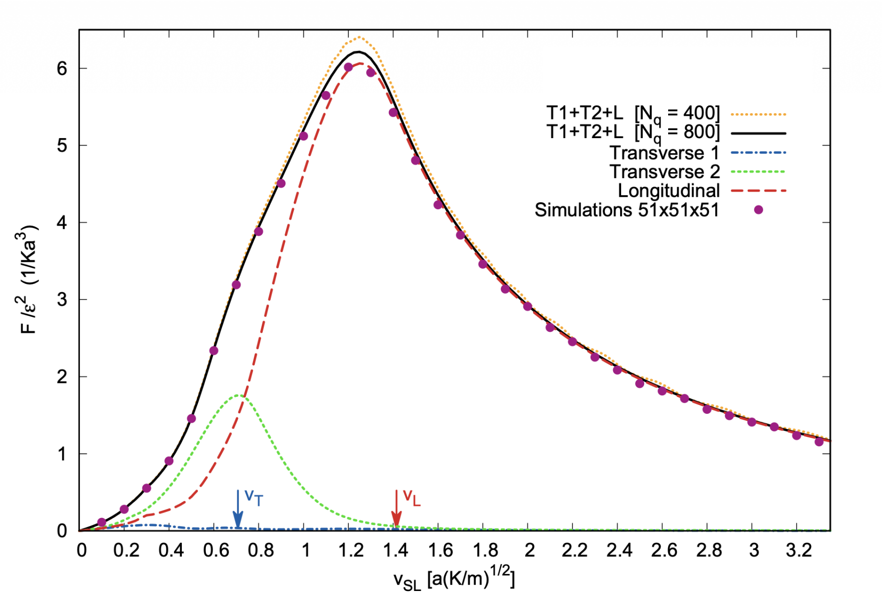

FIGURE 2.4: Friction force as a function of the slider velocity, computed according to Eq. (2.6), with the slider following the line through the cube centers of the sc lattice, as determined by the initial condition $\mathbf{x}_0 = \frac{a}{2}(\mathbf{e}_y + \mathbf{e}_z)$. Black solid line: the total friction. Other curves: the contributions of individual polarization branches. Points: friction evaluated by means of numerical simulations. Arrows: the longitudinal (red) and transverse (blue) speeds of sound in the (100) direction. The reported friction force is divided by ε^2, expressing the strength of the slider-crystal interaction potential. From Ref. [12].

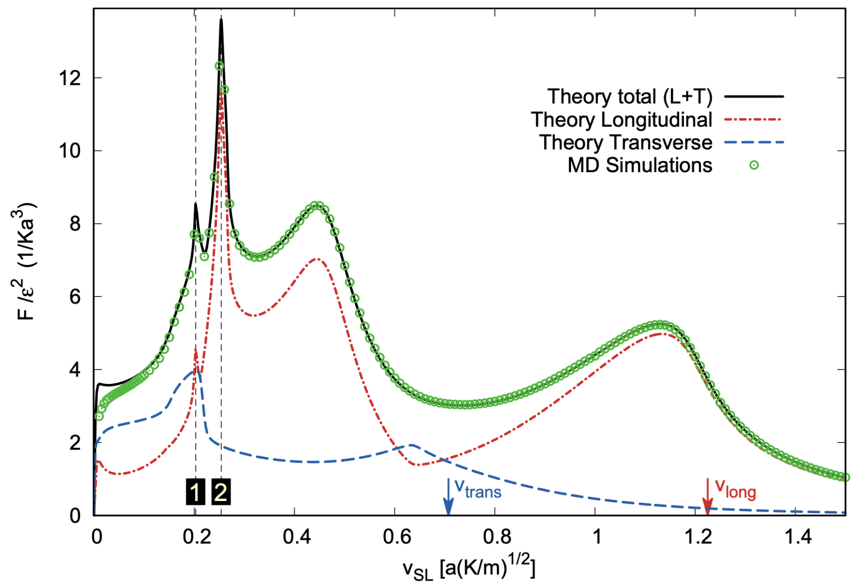

FIGURE 2.5: Comparison of the slider-speed dependence of the friction force F obtained evaluating expression (2.8) (solid curve), with that obtained by numerical MD simulations based on a crystal of 201 × 201 atoms. The slider follows the line through the square centers of the 2D lattice, as determined by the initial condition $\mathbf{x}_0 = \frac{a}{2}\mathbf{e}_y$. Dashed and dot-dashed curves: the contributions to the total friction of the transverse and longitudinal phonons, respectively. Arrows: the longitudinal (red) and transverse (blue) speeds of sound in the (10) direction. From Ref. [12].

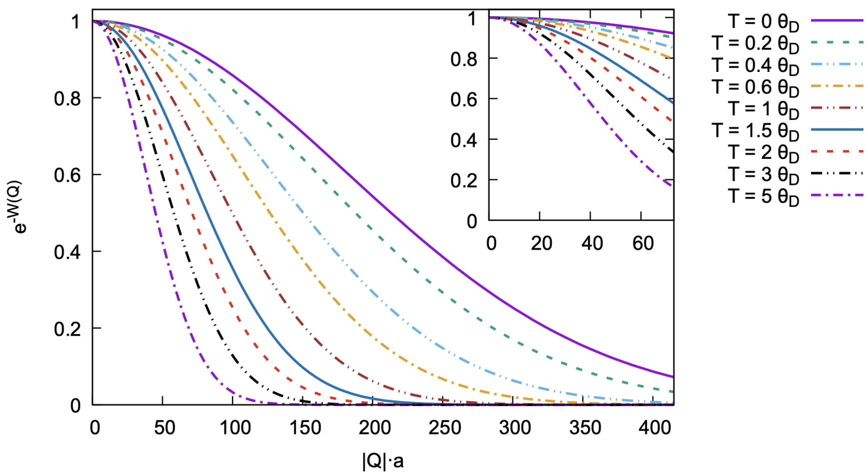

FIGURE 2.6: The Debye-Waller factor for a simple-cubic crystal, as a function of the length of the wave vector **Q** and for a set of temperatures expressed as fractions of the Debye temperature θ_D. The higher the temperature, the faster is the decay of this factor. At the reported physically plausible temperatures, this decay becomes relevant mainly in the high-magnitude **Q** range. From Ref. [13].

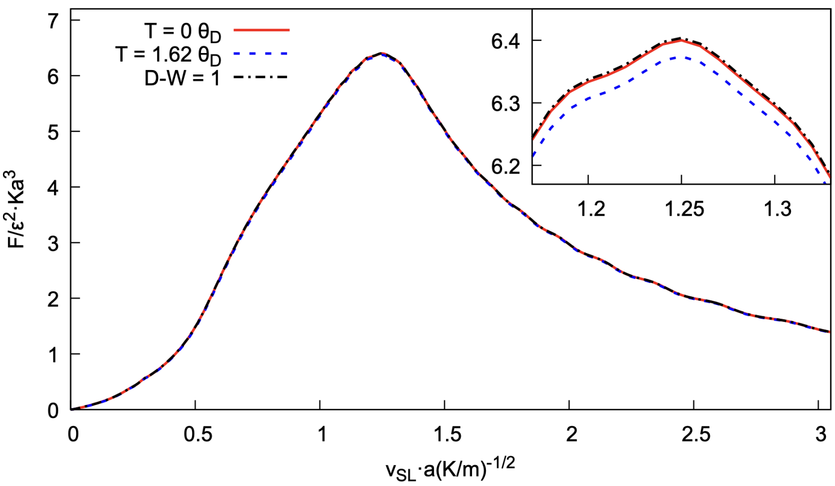

FIGURE 2.7: Comparison of the phonon friction evaluated (i) with the Debye-Waller factors approximated with unity (dot-dashed line); (ii) with the Debye-Waller factors appropriate for $T = 0$ (solid line); and (iii) with the Debye-Waller factors for a finite temperature $T = 1.62\,\theta_D$ (dashed line). Here θ_D is the Debye temperature of the crystal. The physical conditions for the friction results reported here are detailed in Refs. [12, 13].

Chapter 3

Gently grazing a flat crystal surface

In the present chapter we describe in detail the model for frictional sliding over a semi-infinite crystal, and introduce the formalism to evaluate this friction by means of linear-response theory (LRT).

3.1 The model

Consider a 3D harmonic semi-infinite crystal characterized by particles of mass m that at equilibrium are arranged as a simple-cubic lattice truncated at a (001) surface. Harmonic nearest- and second-neighbor springs with elastic constants K and K' and equilibrium lengths a and $\sqrt{2}a$, respectively, guarantee the mechanical stability and determine the phonon dispersions. For the spring constants, we adopt a ratio $K'/K = \frac{1}{2}$ as in Ref. [12]. Neglecting relaxation or reconstruction effects at the surface, we assume that the crystal retains its underlying discrete translational symmetry along the xy-plane.

Crucial ingredients for the evaluation of the response of the semi-infinite crystal are its vibrational frequencies and polarization vectors. Appendix F.3 provides the necessary ingredients to calculate them.

A sliding object (the "slider"), that we represent as a point particle characterized by mass M, position $\mathbf{x}_{\rm SL}$ and velocity $\mathbf{v}_{\rm SL}$, moves over the surface of the crystal. The slider-crystal interaction leads to phonon excitations. In the present work, we do not consider the possibility of the direct excitation of electrons and all kinds of triboelectric phenomena that can occur in real life. The main assumption is that the interaction between the slider and the crystal, which we describe through a 2-body potential, is so weak that it perturbs the slider motion significantly only over very long time scales. For this reason, we can assume that the slider moves at practically constant

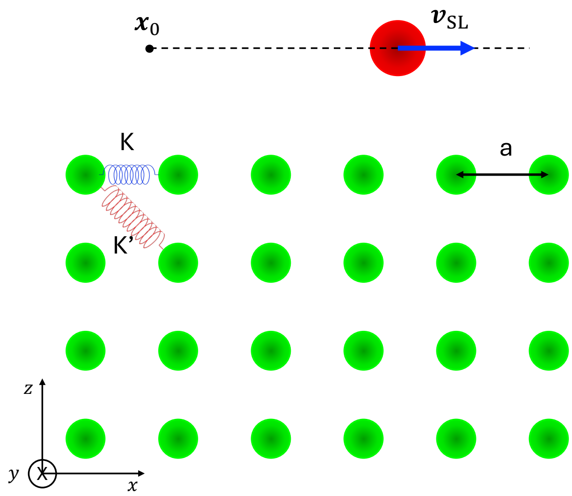

FIGURE 3.1: A sketch of the model: the slider (red sphere) moves over a (001) surface of a simple-cubic crystal, advancing along a (100) high-symmetry line starting off at x_0 (dashed line). The slider interacts with each atom of the semi-infinite crystal through a weak 2-body potential. The vibrational properties of the semi-infinite crystal are determined by first- and second-neighbor springs with elastic constants K and K', respectively.

velocity:

$$\mathbf{x}_{\mathrm{SL}}(t) = \mathbf{x}_0 + \mathbf{v}_{\mathrm{SL}} t \, . \tag{3.1}$$

Here $\mathbf{x}_0$ is the starting point. For the validity of this assumption, the kinetic energy of the particle, $\frac{1}{2} M v_{\mathrm{SL}}^2$, must be much larger than the typical energy transferred in the time taken by the slider to advance by one lattice spacing. Therefore, we need to require not only the interaction strength is small, but also that the slider speed v_{SL} and mass M are sufficiently large. We also suppose that the slider moves along a crystal high-symmetry direction, say (100), namely the x axis: $\mathbf{v}_{\mathrm{SL}} = v_{\mathrm{SL}} \hat{\mathbf{x}}$. Figure 3.1 reports a sketch of the model.

The Hamiltonian of the system can be written as:

$$\begin{aligned} \hat{H} &= \hat{H}_{\mathrm{harm}} + \sum_j \hat{V}(|\hat{\mathbf{x}}_j - \mathbf{x}_{\mathrm{SL}}(t)|) \\ &= \hat{H}_{\mathrm{harm}} + \sum_j \hat{V}(|\hat{\mathbf{x}}_j - \mathbf{x}_0 - \mathbf{v}_{\mathrm{SL}} t|) \, , \end{aligned} \tag{3.2}$$

where $\hat{H}_{\mathrm{harm}}$ is the quantum Hamiltonian for the semi-infinite harmonic lattice, $\hat{V}(r)$ is the 2-body potential energy operator that couples the slider to each atom in the harmonic crystal and $\hat{\mathbf{x}}_j$ is the position operator of the j-th atom. Within the discussed approximation, the slider dynamic becomes

Physical quantity	Natural units	Typical values
length	a	500 pm
mass	m	5×10^{-26} kg
spring constant	K	300 N/m
time	$(m/K)^{1/2}$	1.3×10^{-14} s
angular frequency	$(K/m)^{1/2}$	7.7×10^{13} s^{-1}
velocity	$a(K/m)^{1/2}$	3.9×10^{4} m/s
force	Ka	1.5×10^{-7} N
energy	Ka^2	7.5×10^{-17} J
temperature	Ka^2/k_B	5.4×10^{6} K
action	$a^2(Km)^{1/2}$	9.7×10^{-31} Js

TABLE I: Adopted "natural" units system based on 3 fundamental crystal-related quantities: the lattice spacing a, mass m, and nearest-neighbor spring constant K. We use this system to express all mechanical quantities. We provide indications of plausible values for a standard crystal. Note that the adopted energy scale is of the order of the crystal cohesive energy, and much larger than typical vibrational phonon quanta $\hbar\omega$ or the Debye energy.

trivial, thus the Hamiltonian term for the slider is irrelevant. Introducing the density operator

$$\hat{n}(\mathbf{x}) = \sum_j \delta_3(\mathbf{x} - \hat{\mathbf{x}}_j)\,, \tag{3.3}$$

we can rewrite the Hamiltonian as:

$$\hat{H} = \hat{H}_{\text{harm}} + \int d^3x\, V_{ext}(\mathbf{x}, t)\, \hat{n}(\mathbf{x})\,, \tag{3.4}$$

where

$$V_{ext}(\mathbf{x}, t) \equiv V(|\mathbf{x} - \mathbf{x}_0 - \mathbf{v}_{\text{SL}}t|) = V\left([(\mathbf{x}^{\|} - \mathbf{x}_0^{\|} - \mathbf{v}_{\text{SL}}t)^2 + (z - z_0)^2]^{1/2}\right)\,. \tag{3.5}$$

From now on, the symbol $\|$ stands for the xy components of vectors, namely those parallel to the crystal surface.

This mechanical model is conveniently represented in terms of a system of natural units, based on 3 independent mechanical quantities, which we select to be: the crystal lattice spacing a, mass m, and nearest-neighbor spring constant K. Table I collects all mechanical quantities related to our work in terms of these natural units. It also includes indications of plausible values appropriate for a "standard" crystal.

The hypothesis of a weak coupling between the slider and the crystal allows us to describe friction through the LRT, following an approach that extends that of Chapter 2. We adopt the same regularized LJ potential as in Ref [12]. Appendix B provides the definition of the adopted slider-crystal potential, along with its 2D Fourier transform. Appendix C reports the first and second partial derivatives of the 2D Fourier transform of the regularized LJ potential involved in the Taylor expansion, and consequently in the final friction formula.

Unlike the case of channeling through an infinite cubic crystal, the semi-infinite crystal preserves a discrete symmetry only on the xy-plane, with no symmetry along the (001) direction. Therefore, the reciprocal lattice vectors $\mathbf{G} = 2\pi a^{-1}(l^x, l^y)$, with the (integer) Miller indexes l^x and l^y, are 2D-vectors. Taking advantage of this square-lattice translational invariance, it will be useful to adopt a 2D Fourier representation for the slider-crystal potential (Eq. (3.5)), and for the linear-response function (Eq. (3.7)) introduced in the next section.

3.2 Friction evaluation

Like for the channeling problem, the average friction force F can be evaluated by means of the time-average of the dissipated power by the slider executed over a period $\tau = a/v_{\mathrm{SL}}$, which is the time that the slider takes to advance by an interatomic equilibrium length a, see Eq. (2.3). LRT allows us to calculate the instantaneous power transferred by the slider to the crystal [14]:

$$W = \frac{d}{dt}E(t) \simeq -\int d^3x \int d^3x' \int_{-\infty}^{+\infty} dt'\, V_{ext}(\mathbf{x}, t)\, \frac{\partial \chi^R_{nn}(\mathbf{x}, \mathbf{x}'; t - t')}{\partial t} \times V_{ext}(\mathbf{x}', t')\,. \quad (3.6)$$

Here, $E(t)$ is the internal energy of the crystal at time t, $\chi^R_{nn}(\mathbf{x}, \mathbf{x}'; t-t')$ is the retarded density-density response function of the unperturbed semi-infinite crystal defined as

$$\chi^R_{nn}(\mathbf{x}, \mathbf{x}'; t - t') = -\frac{i}{\hbar}\theta(t - t')\, \langle[\hat{n}(\mathbf{x}, t - t'), \hat{n}(\mathbf{x}', 0)]\rangle\ , \quad (3.7)$$

and $V_{ext}(\mathbf{x}, t)$ is the perturbation produced on the crystal by the slider at time t, see Eq. (3.5).

The bulk of the present project involves precisely the determination of the density-density linear-response function for the semi-infinite harmonic crystal.

Chapter 4

Friction over a surface

In this chapter we provide an analytical expression for friction. For this purpose, we derive an expression for the retarded density-density response function of a semi-infinite dissipative crystal. Using the obtained expression, we calculate an explicit expression for the friction force.

4.1 Dissipation and friction

It is convenient to adopt the following partial Fourier representation for the external potential:

$$V_{ext}(\mathbf{x},t) = \int \frac{d^2q^{\|}}{(2\pi)^2}\, e^{i\mathbf{q}^{\|}\cdot(\mathbf{x}^{\|}-\mathbf{x}_0^{\|}-\mathbf{v}_{\mathrm{SL}}t)}\tilde{V}(\mathbf{q}^{\|},|z-z_0|)\,. \tag{4.1}$$

The 2D Fourier transform of the potential is defined as:

$$\begin{aligned}\tilde{V}(\mathbf{q}^{\|},|z-z_0|) &= \tilde{V}(|\mathbf{q}^{\|}|,|z-z_0|)\\ &= \int d^2r^{\|}\, e^{-i\mathbf{q}^{\|}\cdot\mathbf{r}^{\|}}\, V\left([(\mathbf{r}^{\|})^2+(z-z_0)^2]^{1/2}\right)\,,\end{aligned} \tag{4.2}$$

where

$$\mathbf{r}^{\|} = \mathbf{x}^{\|} - \mathbf{x}_0^{\|} - \mathbf{v}_{\mathrm{SL}}t\,, \tag{4.3}$$

recalling that the slider moves at constant velocity along the x direction ($\mathbf{v}_{\mathrm{SL}} = v_{\mathrm{SL}}\hat{\mathbf{x}}$) following relation (3.1) with $\mathbf{x}_0$ as the starting point.

Taking advantage of the discrete translational invariance along the xy-plane, it is useful to write the 2D inverse Fourier transform of the retarded density-density response function:

$$\begin{aligned}\chi^R_{nn}(\mathbf{x},\mathbf{x}';t-t') = \sum_{\mathbf{G}^{\|}}\int \frac{d^2Q^{\|}}{(2\pi)^2}\int_{-\infty}^{+\infty}\frac{d\omega}{2\pi}e^{-i\omega(t-t')}e^{i\mathbf{Q}^{\|}\cdot\mathbf{x}^{\|}}\times\\ \chi^R_{nn}(\mathbf{Q}^{\|},\mathbf{Q}^{\|}+\mathbf{G}^{\|},z,z';\omega)e^{-i(\mathbf{Q}^{\|}+\mathbf{G}^{\|})\cdot\mathbf{x}'^{\|}}\,,\end{aligned} \tag{4.4}$$

where $\mathbf{G}^{\parallel} = 2\pi a^{-1}(l^x, l^y, 0)$ are the reciprocal lattice vectors of the crystal with Miller indexes l^x, l^y, and of course null off-plane component. The sum over $\mathbf{G}^{\parallel}$ is to be understood as a sum over the integer indexes l^x, l^y. Then, the time derivative of the retarded response function can be written as:

$$\frac{\partial \chi^R_{nn}(\mathbf{x}, \mathbf{x}'; t-t')}{\partial t} = (-i\omega) \sum_{\mathbf{G}^{\parallel}} \int \frac{d^2 Q^{\parallel}}{(2\pi)^2} \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} e^{-i\omega(t-t')} e^{i\mathbf{Q}^{\parallel}\cdot\mathbf{x}^{\parallel}} \times \\ \chi^R_{nn}(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\parallel} + \mathbf{G}^{\parallel}, z, z'; \omega) e^{-i(\mathbf{Q}^{\parallel}+\mathbf{G}^{\parallel})\cdot\mathbf{x}'^{\parallel}} . \quad (4.5)$$

Expressions (3.6), (4.1), (4.5) allow us to derive the time average of the dissipated power executed over a period $\tau = a/v_{\mathrm{SL}}$:

$$\begin{aligned} \bar{W} =& \frac{1}{\tau}\int_0^{\tau} dt\, W(t) \\ =& -\frac{1}{\tau}\int_0^{\tau} dt \int d^3x \int d^3x' \int_{-\infty}^{+\infty} dt'\, V_{ext}(\mathbf{x}, t) \frac{\partial \chi^R_{nn}(\mathbf{x}, \mathbf{x}'; t-t')}{\partial t} \times \\ & V_{ext}(\mathbf{x}', t') \\ =& -\frac{1}{\tau}\int_0^{\tau} dt \int d^3x \int d^3x' \int_{-\infty}^{+\infty} dt' \int \frac{d^2 q^{\parallel}}{(2\pi)^2} \int \frac{d^2 q'^{\parallel}}{(2\pi)^2} \sum_{\mathbf{G}^{\parallel}} \int \frac{d^2 Q^{\parallel}}{(2\pi)^2} \times \\ & \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi}\, e^{i\mathbf{q}^{\parallel}\cdot(\mathbf{x}^{\parallel}-\mathbf{x}_0^{\parallel}-\mathbf{v}_{\mathrm{SL}}t)} e^{i\mathbf{Q}^{\parallel}\cdot\mathbf{x}^{\parallel}} e^{-i\omega(t-t')} \tilde{V}(|\mathbf{q}^{\parallel}|, |z-z_0|)(-i\omega) \times \\ & \chi^R_{nn}(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\parallel}+\mathbf{G}^{\parallel}, z, z'; \omega) e^{i\mathbf{q}'^{\parallel}\cdot(\mathbf{x}'^{\parallel}-\mathbf{x}_0^{\parallel}-\mathbf{v}_{\mathrm{SL}}t')} e^{-i(\mathbf{Q}^{\parallel}+\mathbf{G}^{\parallel})\cdot\mathbf{x}'^{\parallel}} \times \\ & \tilde{V}(|\mathbf{q}'^{\parallel}|, |z'-z_0|) . \end{aligned} \quad (4.6)$$

$V_{ext}(\mathbf{x}', t')$ is real, namely $V_{ext}(\mathbf{x}', t') = V^*_{ext}(\mathbf{x}', t')$:

$$\int \frac{d^2 q^{\parallel}}{(2\pi)^2}\, e^{i\mathbf{q}^{\parallel}\cdot(\mathbf{x}'^{\parallel}-\mathbf{x}_0^{\parallel}-\mathbf{v}_{\mathrm{SL}}t')} \tilde{V}(|\mathbf{q}^{\parallel}|, |z'-z_0|) = \\ \int \frac{d^2 q^{\parallel}}{(2\pi)^2}\, e^{-i\mathbf{q}^{\parallel}\cdot(\mathbf{x}'^{\parallel}-\mathbf{x}_0^{\parallel}-\mathbf{v}_{\mathrm{SL}}t')} \tilde{V}^*(|\mathbf{q}^{\parallel}|, |z'-z_0|) ,$$

applying the transformation $\mathbf{q}^{\parallel} \to -\mathbf{q}^{\parallel}$ to the first integral, we obtain:

$$\int \frac{d^2 q^{\parallel}}{(2\pi)^2}\, e^{-i\mathbf{q}^{\parallel}\cdot(\mathbf{x}'^{\parallel}-\mathbf{x}_0^{\parallel}-\mathbf{v}_{\mathrm{SL}}t')} \tilde{V}(|-\mathbf{q}^{\parallel}|, |z'-z_0|) = \\ \int \frac{d^2 q^{\parallel}}{(2\pi)^2}\, e^{-i\mathbf{q}^{\parallel}\cdot(\mathbf{x}'^{\parallel}-\mathbf{x}_0^{\parallel}-\mathbf{v}_{\mathrm{SL}}t')} \tilde{V}^*(|\mathbf{q}^{\parallel}|, |z'-z_0|) .$$

We observe that

$$\tilde{V}(|-\mathbf{q}^{\parallel}|, |z'-z_0|) = \tilde{V}(|\mathbf{q}^{\parallel}|, |z'-z_0|) = \tilde{V}^*(|\mathbf{q}^{\parallel}|, |z'-z_0|) , \quad (4.7)$$

indicating that the Fourier-transformed function $\tilde{V}$ is real.

Using this property, we can substitute

$$\int \frac{d^2q'^{\parallel}}{(2\pi)^2} e^{i\mathbf{q}'^{\parallel}\cdot(\mathbf{x}'^{\parallel}-\mathbf{x}_0^{\parallel}-\mathbf{v}_{\mathrm{SL}}t')}\tilde{V}(|\mathbf{q}'^{\parallel}|,|z'-z_0|)$$

with

$$\int \frac{d^2q'^{\parallel}}{(2\pi)^2} e^{-i\mathbf{q}'^{\parallel}\cdot(\mathbf{x}'^{\parallel}-\mathbf{x}_0^{\parallel}-\mathbf{v}_{\mathrm{SL}}t')}\tilde{V}(|\mathbf{q}'^{\parallel}|,|z'-z_0|)$$

into equation (4.6) and we obtain:

$$\begin{aligned}
\bar{W} = &-\frac{1}{\tau}\int_0^{\tau} dt \int d^3x \int d^3x' \int_{-\infty}^{+\infty} dt' \int \frac{d^2q^{\parallel}}{(2\pi)^2} \int \frac{d^2q'^{\parallel}}{(2\pi)^2} \sum_{\mathbf{G}^{\parallel}} \int \frac{d^2Q^{\parallel}}{(2\pi)^2}\times \\
&\int_{-\infty}^{+\infty} \frac{d\omega}{2\pi}\, e^{i\mathbf{q}^{\parallel}\cdot(\mathbf{x}^{\parallel}-\mathbf{x}_0^{\parallel}-\mathbf{v}_{\mathrm{SL}}t)} e^{i\mathbf{Q}^{\parallel}\cdot\mathbf{x}^{\parallel}} e^{-i\omega(t-t')}\tilde{V}(|\mathbf{q}^{\parallel}|,|z-z_0|)(-i\omega)\times \\
&\chi^R_{nn}(\mathbf{Q}^{\parallel},\mathbf{Q}^{\parallel}+\mathbf{G}^{\parallel},z,z';\omega) e^{-i\mathbf{q}'^{\parallel}\cdot(\mathbf{x}'^{\parallel}-\mathbf{x}_0^{\parallel}-\mathbf{v}_{\mathrm{SL}}t')} e^{-i(\mathbf{Q}^{\parallel}+\mathbf{G}^{\parallel})\cdot\mathbf{x}'^{\parallel}}\times \\
&\tilde{V}(|\mathbf{q}'^{\parallel}|,|z'-z_0|) \\
= &-\frac{1}{\tau}\int_0^{\tau} dt \int d^3x \int d^3x' \int_{-\infty}^{+\infty} dt' \int \frac{d^2q^{\parallel}}{(2\pi)^2} \int \frac{d^2q'^{\parallel}}{(2\pi)^2} \sum_{\mathbf{G}^{\parallel}} \int \frac{d^2Q^{\parallel}}{(2\pi)^2}\times \\
&\int_{-\infty}^{+\infty} \frac{d\omega}{2\pi}\, e^{i(\mathbf{q}^{\parallel}+\mathbf{Q}^{\parallel})\cdot\mathbf{x}^{\parallel}} e^{-i(\mathbf{q}'^{\parallel}+\mathbf{Q}^{\parallel}+\mathbf{G}^{\parallel})\cdot\mathbf{x}'^{\parallel}} e^{-i\mathbf{q}^{\parallel}\cdot\mathbf{x}_0^{\parallel}} e^{i\mathbf{q}'^{\parallel}\cdot\mathbf{x}_0^{\parallel}}\times \\
&e^{-it(\omega+\mathbf{q}^{\parallel}\cdot\mathbf{v}_{\mathrm{SL}})} e^{it'(\omega+\mathbf{q}'^{\parallel}\cdot\mathbf{v}_{\mathrm{SL}})}(-i\omega)\tilde{V}(|\mathbf{q}^{\parallel}|,|z-z_0|)\times \\
&\chi^R_{nn}(\mathbf{Q}^{\parallel},\mathbf{Q}^{\parallel}+\mathbf{G}^{\parallel},z,z';\omega)\tilde{V}(|\mathbf{q}'^{\parallel}|,|z'-z_0|)\,. \qquad (4.8)
\end{aligned}$$

The integrations over t', $\mathbf{x}^{\parallel}$ and $\mathbf{x}'^{\parallel}$ yield Dirac-δ functions over ω, $\mathbf{q}^{\parallel}$ and $\mathbf{q}'^{\parallel}$, respectively:

$$\int_{-\infty}^{+\infty} dt'\, e^{it'(\omega+\mathbf{q}'^{\parallel}\cdot\mathbf{v}_{\mathrm{SL}})} = 2\pi\delta(\omega+\mathbf{q}'^{\parallel}\cdot\mathbf{v}_{\mathrm{SL}})\,, \qquad (4.9)$$

$$\int d^2x^{\parallel}\, e^{i(\mathbf{q}^{\parallel}+\mathbf{Q}^{\parallel})\cdot\mathbf{x}^{\parallel}} = (2\pi)^2\delta_2(\mathbf{q}^{\parallel}+\mathbf{Q}^{\parallel})\,, \qquad (4.10)$$

$$\int d^2x'^{\parallel}\, e^{-i(\mathbf{q}'^{\parallel}+\mathbf{Q}^{\parallel}+\mathbf{G}^{\parallel})\cdot\mathbf{x}'^{\parallel}} = (2\pi)^2\delta_2(\mathbf{q}'^{\parallel}+\mathbf{Q}^{\parallel}+\mathbf{G}^{\parallel})\,. \qquad (4.11)$$

Accordingly:

$$
\begin{aligned}
\bar{W} =& -\frac{1}{\tau}\int_0^\tau dt \int dz \int dz' \sum_{\mathbf{G}^\parallel} \int \frac{d^2 Q^\parallel}{(2\pi)^2} e^{i\mathbf{Q}^\parallel\cdot\mathbf{x}_0^\parallel} e^{-i(\mathbf{Q}^\parallel+\mathbf{G}^\parallel)\cdot\mathbf{x}_0^\parallel} \times \\
& e^{-it((\mathbf{Q}^\parallel+\mathbf{G}^\parallel)\cdot\mathbf{v}_{\mathrm{SL}}-\mathbf{Q}^\parallel\cdot\mathbf{v}_{\mathrm{SL}})}(-i)(\mathbf{Q}^\parallel+\mathbf{G}^\parallel)\cdot\mathbf{v}_{\mathrm{SL}}\times \\
& \tilde{V}(|\mathbf{Q}^\parallel|,|z-z_0|)\chi^R_{nn}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^\parallel,z,z';(\mathbf{Q}^\parallel+\mathbf{G}^\parallel)\cdot\mathbf{v}_{\mathrm{SL}})\times \\
& \tilde{V}(|\mathbf{Q}^\parallel+\mathbf{G}^\parallel|,|z'-z_0|) \\
=& \frac{i}{\tau}\int_0^\tau dt \int dz \int dz' \sum_{\mathbf{G}^\parallel} e^{-i\mathbf{G}^\parallel\cdot\mathbf{x}_0^\parallel} e^{-it\mathbf{G}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}} \int \frac{d^2Q^\parallel}{(2\pi)^2}(\mathbf{Q}^\parallel+\mathbf{G}^\parallel)\cdot\mathbf{v}_{\mathrm{SL}}\times \\
& \tilde{V}(|\mathbf{Q}^\parallel|,|z-z_0|)\chi^R_{nn}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^\parallel,z,z';(\mathbf{Q}^\parallel+\mathbf{G}^\parallel)\cdot\mathbf{v}_{\mathrm{SL}})\times \\
& \tilde{V}(|\mathbf{Q}^\parallel+\mathbf{G}^\parallel|,|z'-z_0|)\,.
\end{aligned}
\tag{4.12}
$$

Having supposed that the slider moves along the x-direction, the integration over t vanishes whenever $\mathbf{G}^\parallel \cdot \mathbf{v}_{\mathrm{SL}} \neq 0$:

$$
\int_0^\tau dt\, e^{-it\mathbf{G}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}} = \tau\delta(\mathbf{G}^\parallel\cdot\mathbf{v}_{\mathrm{SL}})\,. \tag{4.13}
$$

Indeed, since $\tau = a/v_{\mathrm{SL}}$ and $G^x = \frac{2\pi}{a}l^x$, when $l^x \neq 0$ we obtain:

$$
\begin{aligned}
\int_0^\tau dt\, e^{-it\mathbf{G}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}} &= \left.\frac{e^{-it\mathbf{G}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}}}{-i\mathbf{G}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}}\right|_0^\tau = \frac{e^{-i\tau\mathbf{G}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}}-1}{-i\mathbf{G}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}} \\
&= \frac{e^{-i(a/v_{\mathrm{SL}})2\pi a^{-1}l^x v_{\mathrm{SL}}}-1}{-i\mathbf{G}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}} = \frac{e^{-i2\pi l^x}-1}{-i\mathbf{G}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}} = 0\,.
\end{aligned}
\tag{4.14}
$$

In practice, this leads to restricting the $\mathbf{G}^\parallel$ summation to $\mathbf{G}^y$, just the component of $\mathbf{G}$ perpendicular to $\mathbf{v}_{\mathrm{SL}}$:

$$
\begin{aligned}
\bar{W} = i\int dz \int dz' \sum_{\mathbf{G}^y} e^{-iG^y y_0} \int \frac{d^2Q^\parallel}{(2\pi)^2}\mathbf{Q}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}\times \\
\tilde{V}(|\mathbf{Q}^\parallel|,|z-z_0|)\chi^R_{nn}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,z,z';\mathbf{Q}^\parallel\cdot\mathbf{v}_{\mathrm{SL}})\times \\
\tilde{V}(|\mathbf{Q}^\parallel+\mathbf{G}^y|,|z'-z_0|)\,.
\end{aligned}
\tag{4.15}
$$

The retarded linear-response function satisfies the following symmetry:

$$
\begin{aligned}
&\chi^R_{nn}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,z,z';\mathbf{Q}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}) + \chi^R_{nn}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel-\mathbf{G}^y,z,z';\mathbf{Q}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}) \\
&= [\chi^R_{nn}(-\mathbf{Q}^\parallel,-\mathbf{Q}^\parallel+\mathbf{G}^y,z,z';-\mathbf{Q}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}) \\
&\quad + \chi^R_{nn}(-\mathbf{Q}^\parallel,-\mathbf{Q}^\parallel-\mathbf{G}^y,z,z';-\mathbf{Q}^\parallel\cdot\mathbf{v}_{\mathrm{SL}})]^*.
\end{aligned}
\tag{4.16}
$$

Since $\tilde{V}$ is a real function, we conclude that

$$\begin{aligned}&\chi^R_{nn}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, z, z'; \mathbf{Q}^\parallel \cdot \mathbf{v}_{\mathrm{SL}})\tilde{V}(|\mathbf{Q}^\parallel + \mathbf{G}^y|, |z' - z_0|)\\ &\quad + \chi^R_{nn}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel - \mathbf{G}^y, z, z'; \mathbf{Q}^\parallel \cdot \mathbf{v}_{\mathrm{SL}})\tilde{V}(|\mathbf{Q}^\parallel - \mathbf{G}^y|, |z' - z_0|)\\ &= [\chi^R_{nn}(-\mathbf{Q}^\parallel, -\mathbf{Q}^\parallel + \mathbf{G}^y, z, z'; -\mathbf{Q}^\parallel \cdot \mathbf{v}_{\mathrm{SL}})\tilde{V}(|-\mathbf{Q}^\parallel + \mathbf{G}^y|, |z' - z_0|)\\ &\quad + \chi^R_{nn}(-\mathbf{Q}^\parallel, -\mathbf{Q}^\parallel - \mathbf{G}^y, z, z'; -\mathbf{Q}^\parallel \cdot \mathbf{v}_{\mathrm{SL}})\tilde{V}(|-\mathbf{Q}^\parallel - \mathbf{G}^y|, |z' - z_0|)]^*.\end{aligned} \tag{4.17}$$

This result proves that the real part of

$$\begin{aligned}&\chi^R_{nn}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, z, z'; \mathbf{Q}^\parallel \cdot \mathbf{v}_{\mathrm{SL}})\tilde{V}(|\mathbf{Q}^\parallel + \mathbf{G}^y|, |z' - z_0|)\\ &\qquad + \chi^R_{nn}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel - \mathbf{G}^y, z, z'; \mathbf{Q}^\parallel \cdot \mathbf{v}_{\mathrm{SL}})\tilde{V}(|\mathbf{Q}^\parallel - \mathbf{G}^y|, |z' - z_0|)\end{aligned}$$

is even under the transformation $\mathbf{Q}^\parallel \to -\mathbf{Q}^\parallel$, while its imaginary part is odd. The integrand of Eq. (4.15) can then be written as the product of a $\mathbf{Q}^\parallel$-odd factor, namely $\mathbf{Q}^\parallel \cdot \mathbf{v}_{\mathrm{SL}}$, times the term whose symmetry properties have just been discussed. As the $\mathbf{Q}^\parallel$ integration is carried out over an even domain, only the imaginary part of the retarded linear response function contributes. Introducing the unit vector $\hat{\mathbf{v}}_{\mathrm{SL}} = \mathbf{v}_{\mathrm{SL}}/|\mathbf{v}_{\mathrm{SL}}|$ in slider velocity direction (in practice the $\hat{\mathbf{x}}$ versor), the friction force can then be expressed as:

$$\begin{aligned}F &= \frac{\bar{W}}{v_{\mathrm{SL}}}\\ &= -\int dz \int dz' \sum_{\mathbf{G}^y} e^{-iG^y y_0} \int \frac{d^2Q^\parallel}{(2\pi)^2} \mathbf{Q}^\parallel \cdot \hat{\mathbf{v}}_{\mathrm{SL}} \times\\ &\quad \tilde{V}(|\mathbf{Q}^\parallel|, |z - z_0|) \,\mathrm{Im}\, \chi^R_{nn}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, z, z'; \mathbf{Q}^\parallel \cdot \mathbf{v}_{\mathrm{SL}}) \times\\ &\quad \tilde{V}(|\mathbf{Q}^\parallel + \mathbf{G}^y|, |z' - z_0|)\,.\end{aligned} \tag{4.18}$$

This equation is the first main result of the present thesis, expressing the friction of a weakly-interacting particle grazing the surface of a semi-infinite crystal as a function of the imaginary part of the linear-response function of the crystal itself and the in-plane Fourier-transformed interaction potential energy.

4.2 The retarded linear-response function

We express the retarded linear-response function in its in-plane Fourier representation:

$$\begin{aligned}&\chi^R_{nn}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, z, z'; \omega) = \int d(t - t') e^{i\omega(t-t')} \times\\ &\quad \lim_{S\to\infty} \frac{1}{S} \int_S d^2x^\parallel \int_S d^2x'^\parallel e^{-i\mathbf{Q}^\parallel \cdot \mathbf{x}^\parallel} \chi^R_{nn}(\mathbf{x}, \mathbf{x}', t - t') e^{i(\mathbf{Q}^\parallel + \mathbf{G}^y)\cdot \mathbf{x}'^\parallel}.\end{aligned} \tag{4.19}$$

We rename $t - t' \to t$, therefore:

$$\chi^R_{nn}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, z, z'; \omega) = -\frac{i}{\hbar} \int dt\, \theta(t) e^{i\omega t} \times$$
$$\lim_{S\to\infty} \frac{1}{S} \int_S d^2x^\parallel \int_S d^2x'^\parallel e^{-i\mathbf{Q}^\parallel \cdot \mathbf{x}^\parallel} \Big\langle \Big[\hat{n}(\mathbf{x}, t), \hat{n}(\mathbf{x}', 0)\Big]\Big\rangle e^{i(\mathbf{Q}^\parallel + \mathbf{G}^y)\cdot \mathbf{x}'^\parallel} . \quad (4.20)$$

We introduce the spatial 2D Fourier transform of the density operator:

$$\hat{n}(\mathbf{Q}^\parallel, z, t) = \int_S d^2x^\parallel e^{-i\mathbf{Q}^\parallel\cdot\mathbf{x}^\parallel} \hat{n}(\mathbf{x}, t) \,, \quad (4.21)$$

$$\hat{n}(-\mathbf{Q}^\parallel - \mathbf{G}^y, z', 0) = \int_S d^2x'^\parallel e^{i(\mathbf{Q}^\parallel + \mathbf{G}^y)\cdot\mathbf{x}'^\parallel} \hat{n}(\mathbf{x}', 0) \,. \quad (4.22)$$

Therefore:

$$\chi^R_{nn}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, z, z'; \omega) =$$
$$= -\frac{i}{\hbar} \lim_{S\to\infty} \frac{1}{S} \int_0^\infty dt\; e^{i\omega t} \Big\langle \Big[\hat{n}(\mathbf{Q}^\parallel, z, t), \hat{n}(-\mathbf{Q}^\parallel - \mathbf{G}^y, z', 0)\Big]\Big\rangle . \quad (4.23)$$

We express the position operator of the j-th atom at time t as

$$\hat{\mathbf{x}}_j(t) = \mathbf{R}_j + \hat{\mathbf{u}}_j(t) \,, \quad (4.24)$$

the sum of its equilibrium position $\mathbf{R}_j$ plus its displacement operator $\hat{\mathbf{u}}_j(t)$. We recall the definition of the density operator

$$\hat{n}(\mathbf{x}, t) = \sum_j \delta_3(\mathbf{x} - \hat{\mathbf{x}}_j(t)) \,, \quad (4.25)$$

and by substituting the decomposition (4.24) into Eq. (4.25), we obtain:

$$\hat{n}(\mathbf{x}, t) = \sum_j \delta_2(\mathbf{x}^\parallel - \mathbf{R}^\parallel_j - \hat{\mathbf{u}}^\parallel_j(t))\delta(z - R^z_j - \hat{u}^z_j(t)) \,. \quad (4.26)$$

We note that the displacement operators order evaluated at the same time is arbitrary because they commute as demonstrated in Appendix D, Eq. (D.16).

We substitute expression (4.26) into definition (4.21):

$$\hat{n}(\mathbf{Q}^\parallel, z, t) = \int_S d^2x^\parallel e^{-i\mathbf{Q}^\parallel\cdot\mathbf{x}^\parallel} \sum_j \delta_2(\mathbf{x}^\parallel - \mathbf{R}^\parallel_j - \hat{\mathbf{u}}^\parallel_j(t))\delta(z - R^z_j - \hat{u}^z_j(t))$$
$$= \sum_j e^{-i\mathbf{Q}^\parallel\cdot(\mathbf{R}^\parallel_j + \hat{\mathbf{u}}^\parallel_j(t))}\delta(z - R^z_j - \hat{u}^z_j(t)) \,. \quad (4.27)$$

and likewise for Eq. (4.22).

Then, inserting these results into Eq. (4.23), we obtain:

$$\chi^R_{nn}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, z, z'; \omega) = \\ = -\frac{i}{\hbar} \lim_{S\to\infty} \frac{1}{S} \int_0^\infty dt\, e^{i\omega t} \sum_{j,j'} e^{-i\mathbf{Q}^\parallel \cdot (\mathbf{R}^\parallel_j - \mathbf{R}^\parallel_{j'})} \times \\ \left\langle \left[e^{-i\mathbf{Q}^\parallel \cdot \hat{\mathbf{u}}^\parallel_j(t)} \delta(z - R^z_j - \hat{u}^z_j(t)), e^{i(\mathbf{Q}^\parallel + \mathbf{G}^y)\cdot \hat{\mathbf{u}}^\parallel_{j'}(0)} \delta(z' - R^z_{j'} - \hat{u}^z_{j'}(0)) \right] \right\rangle . \tag{4.28}$$

This expression applies equally well to either a slab of finite thickness or a semi-infinite crystal. In the first case the number of layers counted by the j and j' summations is finite, in the latter case it is infinite.

For a dissipative crystal we introduce a finite phonon lifetime. We add a small imaginary part to the frequency: $\omega \to \omega + i\gamma/2$. This modification introduces a uniform exponential decay with rate $\gamma/2$ to all phonon modes, corresponding to a lifetime $2/\gamma$. With this phenomenological modification, we rewrite the retarded linear response function, Eq. (4.28), as follows:

$$\chi^R_{nn}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, z, z'; \omega) = \\ = -\frac{i}{\hbar} \lim_{S\to\infty} \frac{1}{S} \int_0^\infty dt\, e^{(i\omega - \gamma/2)t} \sum_{j,j'} e^{-i\mathbf{Q}^\parallel \cdot (\mathbf{R}^\parallel_j - \mathbf{R}^\parallel_{j'})} \times \\ \left\langle \left[e^{-i\mathbf{Q}^\parallel \cdot \hat{\mathbf{u}}^\parallel_j(t)} \delta(z - R^z_j - \hat{u}^z_j(t)), e^{i(\mathbf{Q}^\parallel + \mathbf{G}^y)\cdot \hat{\mathbf{u}}^\parallel_{j'}(0)} \delta(z' - R^z_{j'} - \hat{u}^z_{j'}(0)) \right] \right\rangle . \tag{4.29}$$

To calculate the friction force, we need only the imaginary part of χ^R_{nn}, that is:

$$\mathrm{Im}\, \chi^R_{nn}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}_y, z, z'; \omega) = \\ = -\,\mathrm{Re}\Bigg(\frac{1}{\hbar} \lim_{S\to\infty} \frac{1}{S} \int_0^\infty dt\; e^{(i\omega - \gamma/2)t} \sum_{j,j'} e^{-i\mathbf{Q}^\parallel \cdot (\mathbf{R}^\parallel_j - \mathbf{R}^\parallel_{j'})} \times \\ \left\langle \left[e^{-i\mathbf{Q}^\parallel \cdot \hat{\mathbf{u}}^\parallel_j(t)} \delta(z - R^z_j - \hat{u}^z_j(t)), e^{i(\mathbf{Q}^\parallel + \mathbf{G}_y)\cdot \hat{\mathbf{u}}^\parallel_{j'}(0)} \delta(z' - R^z_{j'} - \hat{u}^z_{j'}(0)) \right] \right\rangle \Bigg) . \tag{4.30}$$

4.3 Expression for friction

Now we proceed to derive an analytical expression for the friction force. We start from Eq. (4.18) and we insert the result (4.30) into it:

$$
\begin{aligned}
F = &-\int dz \int dz' \sum_{\mathbf{G}^y} e^{-iG^y y_0} \int \frac{d^2Q^{\|}}{(2\pi)^2} \mathbf{Q}^{\|} \cdot \hat{\mathbf{v}}_{\mathrm{SL}} \times \\
&\tilde{V}(|\mathbf{Q}^{\|}|, |z - z_0|) \, \mathrm{Im}\, \chi^R_{nn}(\mathbf{Q}^{\|}, \mathbf{Q}^{\|} + \mathbf{G}^y, z, z'; \mathbf{Q}^{\|} \cdot \mathbf{v}_{\mathrm{SL}}) \times \\
&\tilde{V}(|\mathbf{Q}^{\|} + \mathbf{G}^y|, |z' - z_0|) \\
= &\int dz \int dz' \sum_{\mathbf{G}^y} e^{-iG^y y_0} \int \frac{d^2Q^{\|}}{(2\pi)^2} \mathbf{Q}^{\|} \cdot \hat{\mathbf{v}}_{\mathrm{SL}} \times \\
&\mathrm{Re}\bigg(\frac{1}{\hbar} \lim_{S \to \infty} \frac{1}{S} \int_0^{\infty} dt\, e^{(i\mathbf{Q}^{\|} \cdot \mathbf{v}_{\mathrm{SL}} - \gamma/2)t} \sum_{j,j'} e^{-i\mathbf{Q}^{\|} \cdot (\mathbf{R}^{\|}_j - \mathbf{R}^{\|}_{j'})} \times \\
&\tilde{V}(|\mathbf{Q}^{\|}|, |z - z_0|) \Big\langle \Big[e^{-i\mathbf{Q}^{\|} \cdot \hat{\mathbf{u}}^{\|}_j(t)} \delta(z - R^z_j - \hat{u}^z_j(t)), \\
&e^{i(\mathbf{Q}^{\|} + \mathbf{G}^y) \cdot \hat{\mathbf{u}}^{\|}_{j'}(0)} \delta(z' - R^z_{j'} - \hat{u}^z_{j'}(0)) \Big] \Big\rangle \tilde{V}(|\mathbf{Q}^{\|} + \mathbf{G}^y|, |z' - z_0|) \bigg) .
\end{aligned} \tag{4.31}
$$

We use definition 1.1 of reference [15] to simplify the integration over z and z':

$$
\begin{aligned}
F = &\lim_{S \to \infty} \frac{1}{S} \sum_{\mathbf{G}^y} e^{-iG^y y_0} \int \frac{d^2Q^{\|}}{(2\pi)^2} \mathbf{Q}^{\|} \cdot \hat{\mathbf{v}}_{\mathrm{SL}} \times \\
&\mathrm{Re}\bigg(\frac{1}{\hbar} \int_0^{\infty} dt\, e^{(i\mathbf{Q}^{\|} \cdot \mathbf{v}_{\mathrm{SL}} - \gamma/2)t} \sum_{j,j'} e^{-i\mathbf{Q}^{\|} \cdot (\mathbf{R}^{\|}_j - \mathbf{R}^{\|}_{j'})} \times \\
&\Big\langle \Big[e^{-i\mathbf{Q}^{\|} \cdot \hat{\mathbf{u}}^{\|}_j(t)} \tilde{V}(|\mathbf{Q}^{\|}|, |R^z_j + \hat{u}^z_j(t) - z_0|), \\
&e^{i(\mathbf{Q}^{\|} + \mathbf{G}^y) \cdot \hat{\mathbf{u}}^{\|}_{j'}(0)} \tilde{V}(|\mathbf{Q}^{\|} + \mathbf{G}^y|, |R^z_{j'} + \hat{u}^z_{j'}(0) - z_0|) \Big] \Big\rangle \bigg) \\
= &\lim_{S \to \infty} \frac{1}{S} \sum_{\mathbf{G}^y} e^{-iG^y y_0} \int \frac{d^2Q^{\|}}{(2\pi)^2} \mathbf{Q}^{\|} \cdot \hat{\mathbf{v}}_{\mathrm{SL}} \times \\
&\mathrm{Re}\bigg(\frac{1}{\hbar} \int_0^{\infty} dt\, e^{(i\mathbf{Q}^{\|} \cdot \mathbf{v}_{\mathrm{SL}} - \gamma/2)t} \sum_{j,j'} e^{-i\mathbf{Q}^{\|} \cdot (\mathbf{R}^{\|}_j - \mathbf{R}^{\|}_{j'})} \times \\
&\Big\langle \Big[e^{-i\mathbf{Q}^{\|} \cdot \hat{\mathbf{u}}^{\|}_j(t)} \tilde{V}(|\mathbf{Q}^{\|}|, z_0 - R^z_j - \hat{u}^z_j(t)), \\
&e^{i(\mathbf{Q}^{\|} + \mathbf{G}^y) \cdot \hat{\mathbf{u}}^{\|}_{j'}(0)} \tilde{V}(|\mathbf{Q}^{\|} + \mathbf{G}^y|, z_0 - R^z_{j'} - \hat{u}^z_{j'}(0)) \Big] \Big\rangle \bigg) .
\end{aligned} \tag{4.32}
$$

In the last expression, we assume that at each instant the probability of finding a particle of the harmonic crystal above the cursor is negligible.

Accordingly we can suppose:

$$z_0 - R_j^z - \hat{u}_j^z(t) > 0\,, \quad z_0 - R_{j'}^z - \hat{u}_{j'}^z(0) > 0 \quad \forall t\,. \tag{4.33}$$

We expand the in-plane Fourier-transformed potential energy relative to its z-displacement variable around the equilibrium separations $z_0 - R_j^z$ in Taylor series truncated at second order:

$$\tilde{V}(|\mathbf{Q}^{\parallel}|, z_0 - R_j^z - \hat{u}_j^z(t)) = \tilde{V}(|\mathbf{Q}^{\parallel}|, z_0 - R_j^z) - \left.\frac{\partial \tilde{V}(|\mathbf{Q}^{\parallel}|, z)}{\partial z}\right|_{z=z_0-R_j^z} \hat{u}_j^z(t) + \frac{1}{2}\left.\frac{\partial^2 \tilde{V}(|\mathbf{Q}^{\parallel}|, z)}{\partial z^2}\right|_{z=z_0-R_j^z} (\hat{u}_j^z(t))^2 + o\big((\hat{u}_j^z(t))^2\big)\,, \tag{4.34}$$

and likewise at $|\mathbf{Q}^{\parallel} + \mathbf{G}^y|$.

We initially focus our attention on the thermal average in Eq. (4.32) substituting the truncated Taylor series of the potential, Eq. (4.34), into it:

$$\begin{aligned}
&\Big\langle \Big[e^{-i\mathbf{Q}^{\parallel}\cdot\hat{\mathbf{u}}_j^{\parallel}(t)} \tilde{V}(|\mathbf{Q}^{\parallel}|, z_0 - R_j^z - \hat{u}_j^z(t)), \\
&e^{i(\mathbf{Q}^{\parallel}+\mathbf{G}^y)\cdot\hat{\mathbf{u}}_{j'}^{\parallel}(0)} \tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z_0 - R_{j'}^z - \hat{u}_{j'}^z(0)) \Big] \Big\rangle \\
&= \Big\langle \Big[e^{-i\mathbf{Q}^{\parallel}\cdot\hat{\mathbf{u}}_j^{\parallel}(t)} \Big(\tilde{V}(|\mathbf{Q}^{\parallel}|, z_0 - R_j^z) - \left.\frac{\partial \tilde{V}(|\mathbf{Q}^{\parallel}|, z)}{\partial z}\right|_{z=z_0-R_j^z} \hat{u}_j^z(t) \\
&\quad + \frac{1}{2}\left.\frac{\partial^2 \tilde{V}(|\mathbf{Q}^{\parallel}|, z)}{\partial z^2}\right|_{z=z_0-R_j^z} (\hat{u}_j^z(t))^2 + o\big((\hat{u}_j^z(t))^2\big) \Big), \\
&\quad e^{i(\mathbf{Q}^{\parallel}+\mathbf{G}^y)\cdot\hat{\mathbf{u}}_{j'}^{\parallel}(0)} \Big(\tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z_0 - R_{j'}^z) - \left.\frac{\partial \tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z')}{\partial z'}\right|_{z'=z_0-R_{j'}^z} \times \\
&\quad \hat{u}_{j'}^z(0) + \frac{1}{2}\left.\frac{\partial^2 \tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z')}{\partial z'^2}\right|_{z'=z_0-R_{j'}^z} (\hat{u}_{j'}^z(0))^2 + o\big((\hat{u}_{j'}^z(0))^2\big) \Big) \Big] \Big\rangle
\end{aligned}$$

$$
\begin{aligned}
&= \tilde{V}(|\mathbf{Q}^{\parallel}|, z_0 - R_j^z)\tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z_0 - R_{j'}^z)\Big\langle\Big[e^{-i\mathbf{Q}^{\parallel}\cdot\hat{\mathbf{u}}_j^{\parallel}(t)}, e^{i(\mathbf{Q}^{\parallel}+\mathbf{G}^y)\cdot\hat{\mathbf{u}}_{j'}^{\parallel}(0)}\Big]\Big\rangle \\
&\quad - \tilde{V}(|\mathbf{Q}^{\parallel}|, z_0 - R_j^z)\frac{\partial\tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z')}{\partial z'}\bigg|_{z'=z_0-R_{j'}^z}\times \\
&\qquad\qquad\qquad \Big\langle\Big[e^{-i\mathbf{Q}^{\parallel}\cdot\hat{\mathbf{u}}_j^{\parallel}(t)}, e^{i(\mathbf{Q}^{\parallel}+\mathbf{G}^y)\cdot\hat{\mathbf{u}}_{j'}^{\parallel}(0)}\hat{u}_{j'}^z(0)\Big]\Big\rangle \\
&\quad - \frac{\partial\tilde{V}(|\mathbf{Q}^{\parallel}|, z)}{\partial z}\bigg|_{z=z_0-R_j^z}\tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z_0 - R_{j'}^z)\times \\
&\qquad\qquad\qquad \Big\langle\Big[e^{-i\mathbf{Q}^{\parallel}\cdot\hat{\mathbf{u}}_j^{\parallel}(t)}\hat{u}_j^z(t), e^{i(\mathbf{Q}^{\parallel}+\mathbf{G}^y)\cdot\hat{\mathbf{u}}_{j'}^{\parallel}(0)}\Big]\Big\rangle \\
&\quad + \frac{1}{2}\tilde{V}(|\mathbf{Q}^{\parallel}|, z_0 - R_j^z)\frac{\partial^2\tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z')}{\partial z'^2}\bigg|_{z'=z_0-R_{j'}^z}\times \\
&\qquad\qquad\qquad \Big\langle\Big[e^{-i\mathbf{Q}^{\parallel}\cdot\hat{\mathbf{u}}_j^{\parallel}(t)}, e^{i(\mathbf{Q}^{\parallel}+\mathbf{G}^y)\cdot\hat{\mathbf{u}}_{j'}^{\parallel}(0)}(\hat{u}_{j'}^z(0))^2\Big]\Big\rangle \\
&\quad + \frac{1}{2}\frac{\partial^2\tilde{V}(|\mathbf{Q}^{\parallel}|, z)}{\partial z^2}\bigg|_{z=z_0-R_j^z}\tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z_0 - R_{j'}^z)\times \\
&\qquad\qquad\qquad \Big\langle\Big[e^{-i\mathbf{Q}^{\parallel}\cdot\hat{\mathbf{u}}_j^{\parallel}(t)}(\hat{u}_j^z(t))^2, e^{i(\mathbf{Q}^{\parallel}+\mathbf{G}^y)\cdot\hat{\mathbf{u}}_{j'}^{\parallel}(0)}\Big]\Big\rangle \\
&\quad + \frac{\partial\tilde{V}(|\mathbf{Q}^{\parallel}|, z)}{\partial z}\bigg|_{z=z_0-R_j^z}\frac{\partial\tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z')}{\partial z'}\bigg|_{z'=z_0-R_{j'}^z}\times \\
&\qquad\qquad\qquad \Big\langle\Big[e^{-i\mathbf{Q}^{\parallel}\cdot\hat{\mathbf{u}}_j^{\parallel}(t)}\hat{u}_j^z(t), e^{i(\mathbf{Q}^{\parallel}+\mathbf{G}^y)\cdot\hat{\mathbf{u}}_{j'}^{\parallel}(0)}\hat{u}_{j'}^z(0)\Big]\Big\rangle \\
&\quad + o\big((\hat{u}^z)^2\big)\,. \qquad (4.35)
\end{aligned}
$$

The rather tedious calculation of the thermal averages in Eq. (4.35) is detailed in Appendix D. In this section, we insert the resulting expression Eq. (D.42) into Eq. (4.32), obtaining the following explicit expression for the friction force:

$$
\begin{aligned}
F = \lim_{S\to\infty}\frac{1}{S}\sum_{\mathbf{G}^y} e^{-iG^y y_0}\int\frac{d^2Q^{\parallel}}{(2\pi)^2}\mathbf{Q}^{\parallel}\cdot\hat{\mathbf{v}}_{\mathrm{SL}}\times \\
\mathrm{Re}\Bigg(\sum_{j,j'} e^{-i\mathbf{Q}^{\parallel}\cdot(\mathbf{R}_j^{\parallel}-\mathbf{R}_{j'}^{\parallel})}e^{-W(\mathbf{Q}^{\parallel},p_j)-W(\mathbf{Q}^{\parallel}+\mathbf{G}^y,p_{j'})}\times \\
\int_0^{\infty} dt\, e^{(i\mathbf{Q}^{\parallel}\cdot\mathbf{v}_{\mathrm{SL}}-\gamma/2)t}\Big(L_{jj'}^{(0)} + L_{jj'}^{(1)} + L_{jj'}^{(2)}\Big)\Bigg)\,, \qquad (4.36)
\end{aligned}
$$

where $L_{jj'}^{(0,1,2)}$ are functions of $(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\parallel} + \mathbf{G}^y, t, z_0, \beta)$ (variables which we will often omit to indicate) describing the effects of lattice vibration up to 2nd order in the out-of-plane displacements $\hat{u}^z$, as we now detail below. The functions $L^{(0,1,2)}_{jj'}$ are dimensionally an Energy·Area^2/Time. $L^{(0)}_{jj'}$ accounts for the zeroth-order term of the potentials expansion:

$$L^{(0)}_{jj'}(\mathbf{Q}^{\|}, \mathbf{Q}^{\|} + \mathbf{G}^y, t, z_0, \beta) = \tilde{V}(|\mathbf{Q}^{\|}|, z_0 - R^z_j)\tilde{V}(|\mathbf{Q}^{\|} + \mathbf{G}^y|, z_0 - R^z_{j'})\times \frac{1}{\hbar}\left(e^{\phi_{jj'}(t,\beta)} - e^{\phi^{\dagger}_{jj'}(t,\beta)}\right). \tag{4.37}$$

$L^{(1)}_{jj'}$ takes the first-order terms of the potentials expansion into account:

$$L^{(1)}_{jj'}(\mathbf{Q}^{\|}, \mathbf{Q}^{\|} + \mathbf{G}^y, t, z_0, \beta) = L^{(A1)}_{jj'} + L^{(B1)}_{jj'}\,, \tag{4.38}$$

where

$$L^{(A1)}_{jj'} = -\tilde{V}(|\mathbf{Q}^{\|}|, z_0 - R^z_j)\frac{\partial \tilde{V}(|\mathbf{Q}^{\|} + \mathbf{G}^y|, z')}{\partial z'}\bigg|_{z'=z_0-R^z_{j'}} \times \frac{i}{\hbar}\Big[e^{\phi_{jj'}(t,\beta)}\Big(2\,W_1(\mathbf{Q}^{\|} + \mathbf{G}^y, p_{j'}) - \phi^{(1a)}_{jj'}(t,\beta)\Big) - e^{\phi^{\dagger}_{jj'}(t,\beta)}\Big(2\,W_1(\mathbf{Q}^{\|} + \mathbf{G}^y, p_{j'}) - \phi^{\dagger(1a)}_{jj'}(t,\beta)\Big)\Big]\,, \tag{4.39}$$

and

$$L^{(B1)}_{jj'} = -\frac{\partial \tilde{V}(|\mathbf{Q}^{\|}|, z)}{\partial z}\bigg|_{z=z_0-R^z_j}\tilde{V}(|\mathbf{Q}^{\|} + \mathbf{G}^y|, z_0 - R^z_{j'}) \times \frac{i}{\hbar}\Big[e^{\phi_{jj'}(t,\beta)}\Big(-2\,W_1(\mathbf{Q}^{\|}, p_j) + \phi^{(1b)}_{jj'}(t,\beta)\Big) - e^{\phi^{\dagger}_{jj'}(t,\beta)}\Big(-2\,W_1(\mathbf{Q}^{\|}, p_j) + \phi^{\dagger(1b)}_{jj'}(t,\beta)\Big)\Big]\,. \tag{4.40}$$

Finally, $L^{(2)}_{jj'}$ accounts for the second-order terms of the potentials expansion:

$$L^{(2)}_{jj'}(\mathbf{Q}^{\|}, \mathbf{Q}^{\|} + \mathbf{G}^y, t, z_0, \beta) = L^{(A2)}_{jj'} + L^{(B2)}_{jj'} + L^{(C2)}_{jj'}\,, \tag{4.41}$$

where

$$L^{(A2)}_{jj'} = \frac{1}{2}\tilde{V}(|\mathbf{Q}^{\|}|, z_0 - R^z_j)\frac{\partial^2 \tilde{V}(|\mathbf{Q}^{\|} + \mathbf{G}^y|, z')}{\partial z'^2}\bigg|_{z'=z_0-R^z_{j'}} \times \frac{1}{\hbar}\Big\{e^{\phi_{jj'}(t,\beta)}\Big[-\Big(2W_1(\mathbf{Q}^{\|} + \mathbf{G}^y, p_{j'}) - \phi^{(1a)}_{jj'}(t,\beta)\Big)^2 + 2W_2(p_{j'})\Big] - e^{\phi^{\dagger}_{jj'}(t,\beta)}\Big[-\Big(2W_1(\mathbf{Q}^{\|} + \mathbf{G}^y, p_{j'}) - \phi^{\dagger(1a)}_{jj'}(t,\beta)\Big)^2 + 2W_2(p_{j'})\Big]\Big\}\,, \tag{4.42}$$

$$L_{jj'}^{(B2)} = \frac{1}{2}\frac{\partial^2 \tilde{V}(|\mathbf{Q}^{\|}|, z)}{\partial z^2}\bigg|_{z=z_0-R_j^z} \tilde{V}(|\mathbf{Q}^{\|} + \mathbf{G}^y|, z_0 - R_{j'}^z) \times$$
$$\frac{1}{\hbar}\left\{ e^{\phi_{jj'}(t,\beta)} \left[-\left(-2W_1(\mathbf{Q}^{\|}, p_j) + \phi_{jj'}^{(1b)}(t,\beta) \right)^2 + 2W_2(p_j) \right] \right.$$
$$\left. - e^{\phi^{\dagger}_{jj'}(t,\beta)} \left[-\left(-2W_1(\mathbf{Q}^{\|}, p_j) + \phi_{jj'}^{\dagger(1b)}(t,\beta) \right)^2 + 2W_2(p_j) \right] \right\}, \quad (4.43)$$

and

$$L_{jj'}^{(C2)} = \frac{\partial \tilde{V}(|\mathbf{Q}^{\|}|, z)}{\partial z}\bigg|_{z=z_0-R_j^z} \frac{\partial \tilde{V}(|\mathbf{Q}^{\|} + \mathbf{G}^y|, z')}{\partial z'}\bigg|_{z'=z_0-R_{j'}^z} \frac{1}{\hbar}\left\{ e^{\phi_{jj'}(t,\beta)} \times \right.$$
$$\left[-\left(-2W_1(\mathbf{Q}^{\|}, p_j) + \phi_{jj'}^{(1b)}(t,\beta) \right)\left(2W_1(\mathbf{Q}^{\|} + \mathbf{G}^y, p_{j'}) - \phi_{jj'}^{(1a)}(t,\beta) \right) \right.$$
$$\left. + \phi_{jj'}^{(2)}(t,\beta) \right] - e^{\phi^{\dagger}_{jj'}(t,\beta)} \left[-\left(-2W_1(\mathbf{Q}^{\|}, p_j) + \phi_{jj'}^{\dagger(1b)}(t,\beta) \right) \times \right.$$
$$\left. \left. \left(2W_1(\mathbf{Q}^{\|} + \mathbf{G}^y, p_{j'}) - \phi_{jj'}^{\dagger(1a)}(t,\beta) \right) + \phi_{jj'}^{\dagger(2)}(t,\beta) \right] \right\}. \quad (4.44)$$

In the previous expressions, the functions $W(\cdot)$, $W_1(\cdot)$, $W_2(\cdot)$ and all the $\phi_{jj'}(\cdot)$, $\phi_{jj'}^{(*)}(\cdot)$, $\phi^{\dagger}_{jj'}(\cdot)$, $\phi_{jj'}^{\dagger(*)}(\cdot)$ are defined in Appendix D from Eq. (D.19) to (D.21), from Eq. (D.24) to (D.27) and from Eq. (D.29) to (D.32), respectively.

Now, to proceed, we need a few important observations:

1. The surface S of the crystal can be written as: $S = N_{xy}\, a^2$ where N_{xy} is the number of particles in the xy-plane and a is the equilibrium spacing between two first-neighbour atoms. Accordingly, the limit in Eq. (4.36) is equivalently attained by sending $N_{xy} \to \infty$.

2. The sum $\sum_{j'}$ over all the atoms of the crystal can be decomposed into two summations: a summation over all the particles of a xy-plane, times a summation over all the planes of the crystal; in symbols: $\sum_{j'} = \sum_{j'_{\|}} \sum_{p_{j'}}$.

3. We proved that all the terms in Eq. (4.36) depend on the horizontal lattice translation $\mathbf{R}_j^{\|} - \mathbf{R}_{j'}^{\|}$ and not individually on $\mathbf{R}_j^{\|}$ and $\mathbf{R}_{j'}^{\|}$; therefore, the sum $\sum_{j'_{\|}}$ yields N_{xy} identical terms.

Using these observations we can rewrite the friction force expression fixing $\mathbf{R}^{\|}_{j'_{\|}=0,\,p_{j'}} = \mathbf{0}$:

$$F = \frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{\mathbf{G}^y} e^{-iG^y y_0} \int \frac{d^2Q^{\|}}{(2\pi)^2} \mathbf{Q}^{\|} \cdot \hat{\mathbf{v}}_{\mathrm{SL}} \times$$
$$\mathrm{Re}\Bigg(\sum_{j,p_{j'}} e^{-i\mathbf{Q}^{\|}\cdot\mathbf{R}^{\|}_j} e^{-W(\mathbf{Q}^{\|},p_j)} e^{-W(\mathbf{Q}^{\|}+\mathbf{G}^y,p_{j'})} \times$$
$$\int_0^\infty dt\, e^{(i\mathbf{Q}^{\|}\cdot\mathbf{v}_{\mathrm{SL}}-\gamma/2)t} \Big(L^{(0)}_{jp_{j'}} + L^{(1)}_{jp_{j'}} + L^{(2)}_{jp_{j'}} \Big) \Bigg) . \tag{4.45}$$

This expression for the friction force involves a sum $\sum_j$ over all the particles of the crystal which makes it quite cumbersome and essentially impossible to compute numerically. To make progress, we adopt the one-phonon approximation for the terms involving $\phi_{jp_{j'}}(t,\beta)$, defined in Eq. (D.24):

$$e^{\phi_{jp_{j'}}(t,\beta)} \simeq 1 + \phi_{jp_{j'}}(t,\beta) \,, \tag{4.46}$$

which is valid in the limit of small $|\phi_{jp_{j'}}(t,\beta)|$, appropriate for not too large wave vector $\mathbf{Q}^{\|}$ and not too high temperature. The analytic expression (4.45) for friction is further processed

1. by rewriting also the sum $\sum_j$ as $\sum_{j_{\|}} \sum_{p_j}$;
2. by executing the thermodynamic limit
$$\lim_{N_{xy}\to\infty} \frac{1}{a^2\, N_{xy}} \sum_{\mathbf{k}}^{BZ} \longrightarrow \int_{BZ} \frac{d^2k}{(2\pi)^2} \,;$$
3. by making use of the 2D version of the periodic delta function, or *Dirac comb*, identity:
$$\sum_{j_{\|}} e^{-i\mathbf{R}^{\|}_j\cdot(\mathbf{Q}^{\|}-\mathbf{k})} = \frac{(2\pi)^2}{a^2} \sum_{\mathbf{G}'} \delta_2(\mathbf{k} - \mathbf{Q}^{\|} - \mathbf{G}') \,.$$

The calculation of the frictional force by means the one-phonon approximation is detailed in Appendix E. Here we report the resulting analytic expression for friction:

$$F = \sum_{\mathbf{G}^y} e^{-iG^y y_0} \int \frac{d^2Q^{\|}}{(2\pi)^2} \mathbf{Q}^{\|} \cdot \hat{\mathbf{v}}_{\mathrm{SL}}\ \mathrm{Re}\Bigg(\sum_{p_j,p_{j'}} e^{-W(\mathbf{Q}^{\|},p_j)} e^{-W(\mathbf{Q}^{\|}+\mathbf{G}^y,p_{j'})} \times$$
$$\int_0^\infty dt\, e^{(i\mathbf{Q}^{\|}\cdot\mathbf{v}_{\mathrm{SL}}-\gamma/2)t} \Big(\mathscr{L}^{(0)}_{p_jp_{j'}} + \mathscr{L}^{(1)}_{p_jp_{j'}} + \mathscr{L}^{(2)}_{p_jp_{j'}} \Big) \Bigg) , \tag{4.47}$$

where $\mathscr{L}^{(k)}_{p_j p_{j'}}$ are functions of $(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\parallel} + \mathbf{G}^y, t, z_0, \beta)$ describing the effects of lattice vibration to k^{th} order in the out-of-plane displacements $\hat{u}^z$, in the one-phonon approximation. The $\mathscr{L}^{(k)}_{p_j p_{j'}}$ functions are dimensionally an Energy·Area/Time.

In particular, $\mathscr{L}^{(0)}_{p_j p_{j'}}$ has the following expression:

$$\begin{aligned}
&\mathscr{L}^{(0)}_{p_j p_{j'}}(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\parallel} + \mathbf{G}^y, t, z_0, \beta) \equiv \\
&\equiv \frac{1}{a^2} \tilde{V}(|\mathbf{Q}^{\parallel}|, z_0 - R^z_{p_j}) \tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z_0 - R^z_{p_{j'}}) \times \\
&\sum_{\lambda} \left[\Phi_{p_j p_{j'}}(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\parallel} + \mathbf{G}^y, \mathbf{Q}^{\parallel}, \lambda, t, \beta) - \Phi^{\dagger}_{p_j p_{j'}}(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\parallel} + \mathbf{G}^y, -\mathbf{Q}^{\parallel}, \lambda, t, \beta) \right],
\end{aligned} \tag{4.48}$$

where $\Phi_{p_j p_{j'}}$ are the following combinations of phonon frequencies, average occupations, and polarization vectors:

$$\begin{aligned}
\Phi_{p_j p_{j'}}(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\parallel} + \mathbf{G}^y, \mathbf{k}, \lambda, t, \beta) &\equiv \frac{1}{2m\omega_\lambda(\mathbf{k})} \mathbf{Q}^{\parallel} \cdot \boldsymbol{\epsilon}_{\lambda, p_j}(\mathbf{k}) \times \\
&(\mathbf{Q}^{\parallel} + \mathbf{G}^y) \cdot \boldsymbol{\epsilon}_{\lambda, p_{j'}}(-\mathbf{k}) [\cos(\omega_\lambda(\mathbf{k})t)(1 + 2n_\lambda(\mathbf{k})) - i \sin(\omega_\lambda(\mathbf{k})t)] \,.
\end{aligned} \tag{4.49}$$

$\Phi_{p_j p_{j'}}$ are dimensionally an (Energy·Time)$^{-1}$.

The more complicated terms $\mathscr{L}^{(1,2)}_{p_j p_{j'}}$ are defined similarly in Appendix E, Eq. (E.20) and (E.21) respectively.

In the zero-temperature limit, both Debye-Waller factors $e^{-W(\mathbf{Q}^{\parallel}, p_j)}$ and the similar one in Eq. (4.45) are expected to be very close to unity, like in Ref. [12]. We observe that in this low-temperature gentle-interaction regime, Planck's constant $\hbar$ disappears from the friction term based on $\mathscr{L}^{(0)}_{p_j p_{j'}}$. Therefore, quantum effects become relevant starting from $\mathscr{L}^{(1)}_{p_j p_{j'}}$. If $p_j = 1$ is the crystal surface layer, $p_j = 2$ is the layer below, and so on, we expect the dominant $\mathscr{L}^{(k)}_{p_j p_{j'}}$ terms to be those associated to small p_j and p'_j. This will happen because the in-plane Fourier transform of the potential and its derivatives decays rapidly away from the crystal surface, see Appendix B, and specifically Figs. B.1, B.2.

Chapter 5

Discussion and conclusions

Equation (4.18) represents the first main result of the present thesis: a general formal expression for the friction of a weakly-interacting particle grazing the surface of a semi-infinite crystal. The friction force results from the integration of the imaginary part of the linear-response function of the crystal itself and the in-plane Fourier-transformed interaction potential energy.

To evaluate this expression we rely on a Taylor expansion of the interaction potential in the phonon displacements $\hat{u}^z$ in the direction perpendicular to the surface. Adding to the truncation of this expansion a few further approximations, e.g. the one-phonon approximation, we obtain the approximate formula (4.47). $\mathscr{L}^{(k)}_{p_j p_{j'}}$ terms involve the in-plane 2D Fourier transform of the potential, and its derivatives with respect to the z coordinate perpendicular to the surface. In Appendix C we provide an analytic expressions of such derivatives for the regularized LJ potential defined in Appendix B. Another crucial ingredient of $\mathscr{L}^{(k)}_{p_j p_{j'}}$ is the determination of the phonon frequencies and polarization vectors. One can proceed in two alternative ways: by diagonalizing the dynamical matrix for a sufficiently thick crystal slab along the z-direction (whose elements are calculated in Appendix F.3), or by the Green's function method for a semi-infinite crystal. In Appendix G we propose a method to evaluate, by means the Green's functions, the normal modes for a semi-infinite 1D chain, but it needs to be extended to the 3D semi-infinite crystal [3].

To continue this research, the next step is to write a code for the numerical evaluation of Eq. (4.47): the computationally complex part will be related to calculate $\mathscr{L}^{(k)}_{p_j p_{j'}}$ terms. At that point it will be possible to compare these analytic results with numerical simulation and possibly even experimental techniques.

Further new challenges will be to extend the present theory to crystals with different lattice structures, such as fcc, bcc, hcp, etc. Another relevant extension will involve taking any relaxation or reconstruction effects at the surface into account.

Appendix A

Linear response theory

LRT studies how a system in thermal equilibrium is modified by the introduction of a weak external perturbation. We add to the unperturbed Hamiltonian $\hat{H}_0$ a time-dependent term $\hat{V}(t)$ of the general form:

$$\hat{V}(t) = \int d^3x\, \hat{A}(\mathbf{x})\, \varphi(\mathbf{x}, t)\,, \tag{A.1}$$

where $\varphi(\mathbf{x}, t)$ is an external perturbation which couples with the Hermitian operator $\hat{A}(\mathbf{x})$.

The thermal average of a second operator $\hat{B}(\mathbf{x})$ is defined as:

$$\langle \hat{B}(\mathbf{x}, t)\rangle \equiv \mathrm{Tr}[\hat{\rho}(t)\hat{B}(\mathbf{x})]\,, \tag{A.2}$$

where $\hat{\rho}(t)$ is the time-dependent density matrix operator of the perturbed system. LRT allows us to calculate $\langle \hat{B}(\mathbf{x}, t)\rangle$ at first order in the perturbation. Introducing the interaction picture of the density matrix operator and expanding it at first perturbative order one can demonstrate:

$$\begin{aligned}\langle \hat{B}(\mathbf{x}, t)\rangle \simeq \langle \hat{B}(\mathbf{x})\rangle_0 - \frac{i}{\hbar}\int_{-\infty}^{+\infty} dt' \int d^3x'\, \theta(t-t') \times \\ \mathrm{Tr}(\hat{\rho}_0[\hat{B}(\mathbf{x}, t), \hat{A}(\mathbf{x}', t')])\varphi(\mathbf{x}', t')\,,\end{aligned} \tag{A.3}$$

where $\langle \hat{B}(\mathbf{x})\rangle_0 = \mathrm{Tr}[\hat{\rho}_0 \hat{B}(\mathbf{x})]$ with $\hat{\rho}_0 = \frac{e^{-\beta \hat{H}_0}}{Z_0}$ and Z_0 is the partition function of the unperturbed system.

The retarded linear response function is defined as:

$$\begin{aligned}\chi^R_{BA}(\mathbf{x}, \mathbf{x}', t, t') &\equiv -\frac{i}{\hbar}\theta(t-t')\mathrm{Tr}(\hat{\rho}_0[\hat{B}(\mathbf{x}, t), \hat{A}(\mathbf{x}', t')]) \\ &= -\frac{i}{\hbar}\theta(t-t')\mathrm{Tr}(\hat{\rho}_0[\hat{B}(\mathbf{x}, t-t'), \hat{A}(\mathbf{x}')]) \\ &= -\frac{i}{\hbar}\theta(t-t')\langle[\hat{B}(\mathbf{x}, t-t'), \hat{A}(\mathbf{x}')]\rangle \equiv \chi^R_{BA}(\mathbf{x}, \mathbf{x}', t-t')\,,\end{aligned} \tag{A.4}$$

where $\langle ... \rangle$ indicates the thermal average with the unperturbed density matrix. It is easy to show that retarded linear response function depends only on the difference in time. Therefore, one has all the elements to write the LRT expression for $\langle \hat{B}(\mathbf{x}, t) \rangle$:

$$\langle \hat{B}(\mathbf{x}, t) \rangle \simeq \langle \hat{B}(\mathbf{x}) \rangle_0 + \int_{-\infty}^{+\infty} dt' \int d^3x' \, \chi^R_{BA}(\mathbf{x}, \mathbf{x}', t - t') \varphi(\mathbf{x}', t') \, . \qquad \text{(A.5)}$$

For more information, see Refs. [16, 17, 18].

Appendix B

2D Fourier Transform of the potential

The 2D Fourier transform of the interaction potential is:

$$\begin{aligned}\tilde{V}(|\mathbf{q}_{\|}|, |z - z_0|) &= \int d^2 r_{\|} e^{-i\mathbf{q}_{\|}\cdot\mathbf{r}_{\|}} V(\sqrt{r_{\|}^2 + (z - z_0)^2}) \\ &= \int_0^{\infty} dr_{\|} r_{\|} V(\sqrt{r_{\|}^2 + (z - z_0)^2}) \int_0^{2\pi} d\theta e^{-iq_{\|} r_{\|} \cos\theta} . \end{aligned} \quad \text{(B.1)}$$

For simplicity of notation, we evaluate this expression for z in place of $|z - z_0|$:

$$\tilde{V}(q_{\|}, z) = \int_0^{\infty} dr_{\|} r_{\|} V(\sqrt{r_{\|}^2 + z^2}) \int_0^{2\pi} d\theta e^{-iq_{\|} r_{\|} \cos\theta} . \quad \text{(B.2)}$$

We remember the definition of n^{th}-order Bessel function of the first kind:

$$J_n(x) = \frac{1}{2\pi i^n} \int_0^{2\pi} e^{ix\cos\theta} e^{in\theta} d\theta . \quad \text{(B.3)}$$

For our purpose we use $n = 0$, therefore:

$$J_0(x) = \frac{1}{2\pi} \int_0^{2\pi} e^{ix\cos\theta} d\theta . \quad \text{(B.4)}$$

We can observe that $J_0(x)$ is even, that is

$$J_0(x) = J_0(-x) . \quad \text{(B.5)}$$

Using the relations (B.4) and (B.5), we solve the equation (B.2):

$$\tilde{V}(q_{\|}, z) = 2\pi \int_0^{\infty} dr_{\|} r_{\|} V(\sqrt{r_{\|}^2 + z^2}) J_0(q_{\|} r_{\|}) . \quad \text{(B.6)}$$

The relation (B.6) is valid for any choice of external potential.

We need a weak potential to make the perturbative approach meaningful; therefore, we adopt a potential that remains finite even at close range. Among the various possible potentials, we decide to use a regularized Lennard-Jones potential, as proposed in the reference [12], defined by:

$$V(\sqrt{r_{\parallel}^2 + z^2}) = \varepsilon \left[\left(\frac{\sigma^2 + d^2}{r_{\parallel}^2 + z^2 + d^2} \right)^6 - 2 \left(\frac{\sigma^2 + d^2}{r_{\parallel}^2 + z^2 + d^2} \right)^3 \right] . \tag{B.7}$$

We insert the definition (B.7) into the equation (B.6):

$$\begin{aligned} \tilde{V}(q_{\parallel}, z) =& 2\pi\varepsilon \int_0^\infty dr_{\parallel} r_{\parallel} \times \\ & \left[\left(\frac{\sigma^2 + d^2}{r_{\parallel}^2 + z^2 + d^2} \right)^6 - 2 \left(\frac{\sigma^2 + d^2}{r_{\parallel}^2 + z^2 + d^2} \right)^3 \right] J_0(q_{\parallel} r_{\parallel}) \,. \end{aligned} \tag{B.8}$$

To derive an explicit expression for the integral in Eq. (B.8), we resort to equation 10.22.46 of Ref. [19]:

$$\int_0^\infty \frac{t^{\nu+1} J_\nu(at)}{(t^2 + b^2)^{\mu+1}} dt = \frac{a^\mu b^{\nu-\mu}}{2^\mu \, \Gamma(\mu+1)} K_{|\nu-\mu|}(ab) \,, \tag{B.9}$$

where $K_{|\nu-\mu|}(\cdot)$ is a modified Bessel function of the second kind and $\Gamma(\cdot)$ is the Euler's Gamma. For our case of interest we specialize the result (B.9) to $\nu = 0$:

$$\int_0^\infty \frac{t \, J_0(at)}{(t^2 + b^2)^{\mu+1}} dt = \frac{a^\mu b^{-\mu}}{2^\mu \, \Gamma(\mu+1)} K_{|-\mu|}(ab) \,. \tag{B.10}$$

By applying this formula twice, we solve the equation (B.8):

$$\begin{aligned} \tilde{V}(q_{\parallel}, z) =& 2\pi\varepsilon(\sigma^2 + d^2)^3 \left[(\sigma^2 + d^2)^3 \int_0^\infty dr_{\parallel} r_{\parallel} \frac{1}{(r_{\parallel}^2 + z^2 + d^2)^6} J_0(q_{\parallel} r_{\parallel}) \right. \\ & \left. -2 \int_0^\infty dr_{\parallel} r_{\parallel} \frac{1}{(r_{\parallel}^2 + z^2 + d^2)^3} J_0(q_{\parallel} r_{\parallel}) \right] \\ =& 2\pi\varepsilon(\sigma^2 + d^2)^3 \left[\frac{(\sigma^2 + d^2)^3}{2^5 \, 5!} \frac{q_{\parallel}^5}{(\sqrt{z^2 + d^2})^5} K_5(q_{\parallel} \sqrt{z^2 + d^2}) \right. \\ & \left. - \frac{2}{2^2 \, 2!} \frac{q_{\parallel}^2}{z^2 + d^2} K_2(q_{\parallel} \sqrt{z^2 + d^2}) \right] , \end{aligned} \tag{B.11}$$

where we considered $a = q_{\parallel}$, $b = \sqrt{z^2 + d^2}$ and we use the property:

$$\Gamma(n + 1) = n! \tag{B.12}$$

for all natural numbers n.

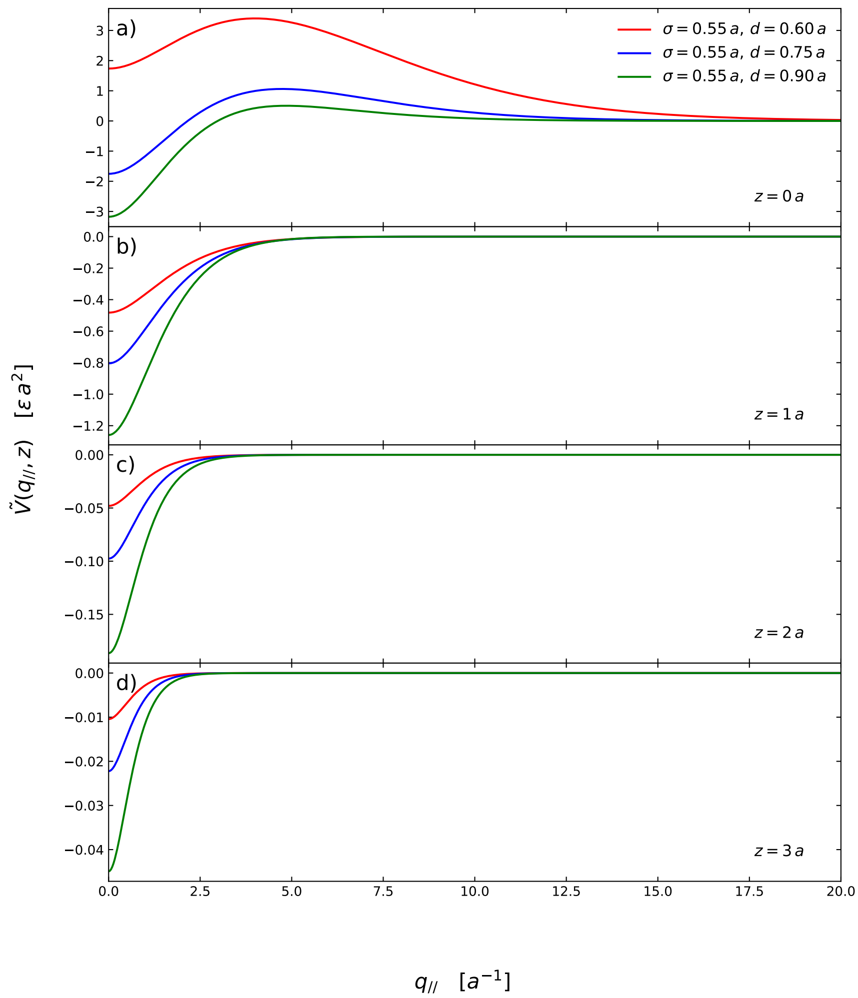

FIGURE B.1: 2D Fourier Transform of the regularized LJ potential $\tilde{V}(q_\parallel, z)$, Eq. (B.11), for the same values of σ and d and different z-values.

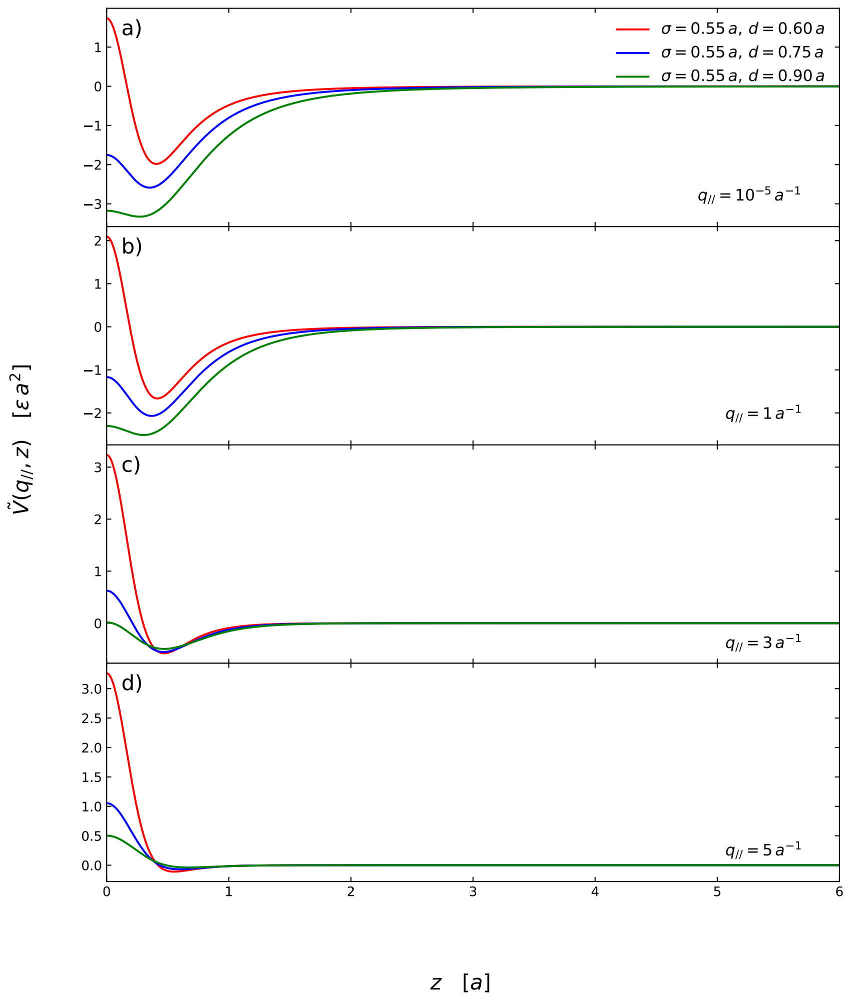

FIGURE B.2: 2D Fourier Transform of the regularized LJ potential $\tilde{V}(q_{\|}, z)$, Eq. (B.11), for the same values of σ and d and different $q_{\|}$-values.

Appendix C

First and second partial derivatives of the 2D Fourier Transform of the potential

We want to calculate the first and second derivatives of the 2D Fourier Transform of the potential chosen in Appendix B: a regularized Lennard-Jones potential. We rewrite it for convenience:

$$V(\sqrt{r_{\parallel}^2 + z^2}) = \varepsilon \left[\left(\frac{\sigma^2 + d^2}{r_{\parallel}^2 + z^2 + d^2} \right)^6 - 2 \left(\frac{\sigma^2 + d^2}{r_{\parallel}^2 + z^2 + d^2} \right)^3 \right] . \tag{C.1}$$

Let us start by evaluating the first partial derivative of this potential (C.1):

$$\begin{aligned}
&\frac{\partial V(\sqrt{r_{\parallel}^2 + z^2})}{\partial z} \\
&= \varepsilon \left[(\sigma^2 + d^2)^6 (-6) \frac{2z}{(r_{\parallel}^2 + z^2 + d^2)^7} - 2(\sigma^2 + d^2)^3 (-3) \frac{2z}{(r_{\parallel}^2 + z^2 + d^2)^4} \right] \\
&= 12\varepsilon (\sigma^2 + d^2)^3 z \left[-(\sigma^2 + d^2)^3 \frac{1}{(r_{\parallel}^2 + z^2 + d^2)^7} + \frac{1}{(r_{\parallel}^2 + z^2 + d^2)^4} \right] .
\end{aligned} \tag{C.2}$$

The 2D Fourier Transform of $\frac{\partial V(\sqrt{r_{\parallel}^2 + z^2})}{\partial z}$ is:

$$\begin{aligned}
\frac{\partial \tilde{V}(q_{\parallel}, z)}{\partial z} &= \int_0^{\infty} dr_{\parallel} r_{\parallel} \frac{\partial V(\sqrt{r_{\parallel}^2 + z^2})}{\partial z} \int_0^{2\pi} d\theta e^{-iq_{\parallel} r_{\parallel} \cos(\theta)} \\
&= 2\pi \int_0^{\infty} dr_{\parallel} r_{\parallel} \frac{\partial V(\sqrt{r_{\parallel}^2 + z^2})}{\partial z} J_0(q_{\parallel} r_{\parallel}) \, ,
\end{aligned} \tag{C.3}$$

where $J_0(q_\parallel r_\parallel)$ is defined by Eq. (B.4).

Now, we substitute the expression (C.2) into Eq. (C.3):

$$\begin{aligned}\frac{\partial \tilde{V}(q_\parallel, z)}{\partial z} =& 24\pi\varepsilon(\sigma^2+d^2)^3 z\Bigg[-(\sigma^2+d^2)^3\int_0^\infty dr_\parallel r_\parallel \frac{J_0(q_\parallel r_\parallel)}{(r_\parallel^2+z^2+d^2)^7} \\ &+ \int_0^\infty dr_\parallel r_\parallel \frac{J_0(q_\parallel r_\parallel)}{(r_\parallel^2+z^2+d^2)^4}\Bigg] \\ =& 24\pi\varepsilon(\sigma^2+d^2)^3 z\Bigg[-\frac{(\sigma^2+d^2)^3}{2^6\,6!}\frac{q_\parallel^6}{(z^2+d^2)^3}K_6(q_\parallel\sqrt{z^2+d^2}) \\ &+ \frac{1}{2^3\,3!}\frac{q_\parallel^3}{(\sqrt{z^2+d^2})^3}K_3(q_\parallel\sqrt{z^2+d^2})\Bigg],\end{aligned} \tag{C.4}$$

where we applied the results (B.10) and (B.12).

The second partial derivative of the potential (C.1) is

$$\begin{aligned}&\frac{\partial^2 V(\sqrt{r_\parallel^2+z^2})}{\partial z^2} \\ &= 12\varepsilon(\sigma^2+d^2)^3\Bigg[-(\sigma^2+d^2)^3\frac{1}{(r_\parallel^2+z^2+d^2)^7}+\frac{1}{(r_\parallel^2+z^2+d^2)^4}\Bigg] \\ &+ 12\varepsilon(\sigma^2+d^2)^3 z\Bigg[-(\sigma^2+d^2)^3\frac{-14z}{(r_\parallel^2+z^2+d^2)^8}-\frac{8z}{(r_\parallel^2+z^2+d^2)^5}\Bigg] \\ &= 12\varepsilon(\sigma^2+d^2)^3\Bigg[-(\sigma^2+d^2)^3\frac{1}{(r_\parallel^2+z^2+d^2)^7}+\frac{1}{(r_\parallel^2+z^2+d^2)^4} \\ &+ 14z^2(\sigma^2+d^2)^3\frac{1}{(r_\parallel^2+z^2+d^2)^8}-8z^2\frac{1}{(r_\parallel^2+z^2+d^2)^5}\Bigg].\end{aligned} \tag{C.5}$$

The 2D Fourier Transform of $\frac{\partial^2 V(\sqrt{r_\parallel^2+z^2})}{\partial z^2}$ is:

$$\begin{aligned}\frac{\partial^2 \tilde{V}(q_\parallel, z)}{\partial z^2} =& \int_0^\infty dr_\parallel r_\parallel \frac{\partial^2 V(\sqrt{r_\parallel^2+z^2})}{\partial z^2}\int_0^{2\pi} d\theta e^{-iq_\parallel r_\parallel\cos(\theta)} \\ =& 2\pi\int_0^\infty dr_\parallel r_\parallel \frac{\partial^2 V(\sqrt{r_\parallel^2+z^2})}{\partial z^2}J_0(q_\parallel r_\parallel),\end{aligned} \tag{C.6}$$

where $J_0(q_\parallel r_\parallel)$ is defined by Eq. (B.4).

Now, we substitute the expression (C.5) into Eq. (C.6):

$$\frac{\partial^2 \tilde{V}(q_\parallel, z)}{\partial z^2} = 24\pi\varepsilon(\sigma^2+d^2)^3\Bigg[-(\sigma^2+d^2)^3\int_0^\infty dr_\parallel r_\parallel \frac{J_0(q_\parallel r_\parallel)}{(r_\parallel^2+z^2+d^2)^7} + \int_0^\infty dr_\parallel r_\parallel \frac{J_0(q_\parallel r_\parallel)}{(r_\parallel^2+z^2+d^2)^4} + 14z^2(\sigma^2+d^2)^3 \times \int_0^\infty dr_\parallel r_\parallel \frac{J_0(q_\parallel r_\parallel)}{(r_\parallel^2+z^2+d^2)^8} - 8z^2\int_0^\infty dr_\parallel r_\parallel \frac{J_0(q_\parallel r_\parallel)}{(r_\parallel^2+z^2+d^2)^5}\Bigg]. \tag{C.7}$$

By applying the results (B.10) and (B.12), we solve the equation:

$$\begin{aligned}\frac{\partial^2 \tilde{V}(q_\parallel, z)}{\partial z^2} =& 24\pi\varepsilon(\sigma^2+d^2)^3\Bigg[-\frac{(\sigma^2+d^2)^3}{2^6\,6!}\frac{q_\parallel^6}{(z^2+d^2)^3}K_6(q_\parallel\sqrt{z^2+d^2}) \\ &+ \frac{1}{2^3\,3!}\frac{q_\parallel^3}{(\sqrt{z^2+d^2})^3}K_3(q_\parallel\sqrt{z^2+d^2}) + 14z^2\frac{(\sigma^2+d^2)^3}{2^7\,7!}\times \\ &\frac{q_\parallel^7}{(\sqrt{z^2+d^2})^7}K_7(q_\parallel\sqrt{z^2+d^2}) - 8z^2\frac{1}{2^4\,4!}\frac{q_\parallel^4}{(z^2+d^2)^2}\times \\ &K_4(q_\parallel\sqrt{z^2+d^2})\Bigg] \\ =& 24\pi\varepsilon(\sigma^2+d^2)^3\Bigg[\frac{(\sigma^2+d^2)^3}{2^6\,6!}\frac{q_\parallel^6}{(z^2+d^2)^3}\Bigg(-K_6(q_\parallel\sqrt{z^2+d^2}) \\ &+ z^2\frac{q_\parallel}{\sqrt{z^2+d^2}}K_7(q_\parallel\sqrt{z^2+d^2})\Bigg) + \frac{1}{2^3\,3!}\frac{q_\parallel^3}{(\sqrt{z^2+d^2})^3}\times \\ &\Bigg(K_3(q_\parallel\sqrt{z^2+d^2}) - z^2\frac{q_\parallel}{\sqrt{z^2+d^2}}K_4(q_\parallel\sqrt{z^2+d^2})\Bigg)\Bigg].\end{aligned} \tag{C.8}$$

Appendix D

Thermal average: explicit derivation

We want to provide the detailed calculation of the thermal average expression (4.35). For convenience, let us rewrite it:

$$
\begin{aligned}
&\langle [e^{-i\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t)}\tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz} - \hat{u}_{jz}(t)), e^{i(\mathbf{Q}_\parallel+\mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0)}\times \\
&\tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z_0 - R_{j'z} - \hat{u}_{j'z}(0))]\rangle \\
&\simeq \tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz})\tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z_0 - R_{j'z})\times \\
&\langle [e^{-i\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t)}, e^{i(\mathbf{Q}_\parallel+\mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0)}]\rangle \\
&-\tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz})\frac{\partial\tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z')}{\partial z'}\bigg|_{z'=z_0-R_{j'z}}\times \\
&\langle [e^{-i\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t)}, e^{i(\mathbf{Q}_\parallel+\mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0)}\hat{u}_{j'z}(0)]\rangle \\
&-\frac{\partial\tilde{V}(|\mathbf{Q}_\parallel|, z)}{\partial z}\bigg|_{z=z_0-R_{jz}}\tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z_0 - R_{j'z})\times \\
&\langle [e^{-i\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t)}\hat{u}_{jz}(t), e^{i(\mathbf{Q}_\parallel+\mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0)}]\rangle \\
&+\frac{1}{2}\tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz})\frac{\partial^2\tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z')}{\partial z'^2}\bigg|_{z'=z_0-R_{j'z}}\times \\
&\langle [e^{-i\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t)}, e^{i(\mathbf{Q}_\parallel+\mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0)}(\hat{u}_{j'z}(0))^2]\rangle \\
&+\frac{1}{2}\frac{\partial^2\tilde{V}(|\mathbf{Q}_\parallel|, z)}{\partial z^2}\bigg|_{z=z_0-R_{jz}}\tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z_0 - R_{j'z})\times \\
&\langle [e^{-i\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t)}(\hat{u}_{jz}(t))^2, e^{i(\mathbf{Q}_\parallel+\mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0)}]\rangle \\
&+\frac{\partial\tilde{V}(|\mathbf{Q}_\parallel|, z)}{\partial z}\bigg|_{z=z_0-R_{jz}}\frac{\partial\tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z')}{\partial z'}\bigg|_{z'=z_0-R_{j'z}}\times \\
&\langle [e^{-i\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t)}\hat{u}_{jz}(t), e^{i(\mathbf{Q}_\parallel+\mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0)}\hat{u}_{j'z}(0)]\rangle
\end{aligned}
$$

$$\equiv A0 - A1 - B1 + A2 + B2 + C2\,. \tag{D.1}$$

We observe that each term of the relation (D.1) comes from only two generating functions:

$$J(\xi_j, \xi'_{j'}) = \langle e^{-i\mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t) + \xi_j \hat{u}_{jz}(t)} e^{i(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot \hat{\mathbf{u}}_{j'\parallel}(0) + \xi'_{j'} \hat{u}_{j'z}(0)} \rangle\,, \tag{D.2}$$

$$J'(\xi_j, \xi'_{j'}) = \langle e^{i(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot \hat{\mathbf{u}}_{j'\parallel}(0) + \xi'_{j'} \hat{u}_{j'z}(0)} e^{-i\mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t) + \xi_j \hat{u}_{jz}(t)} \rangle\,. \tag{D.3}$$

To evaluate them, we use the Gaussian identity:

$$\langle e^A e^B \rangle = e^{\frac{1}{2}\langle A^2\rangle + \frac{1}{2}\langle B^2 \rangle + \langle A\,B\rangle} \tag{D.4}$$

valid for harmonic-oscillator operators [20], calling

$$A = -i\mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t) + \xi_j \hat{u}_{jz}(t)\,, \tag{D.5}$$

$$B = i(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot \hat{\mathbf{u}}_{j'\parallel}(0) + \xi'_{j'} \hat{u}_{j'z}(0)\,. \tag{D.6}$$

To evaluate $\langle A^2 \rangle$, $\langle B^2 \rangle$, $\langle AB \rangle$ and $\langle BA \rangle$, we express the displacement operators in terms of phonon annihilation $\hat{b}_{\mathbf{k}\lambda}$ and creation $\hat{b}^\dagger_{\mathbf{k}\lambda}$ operators [21]:

$$\hat{\mathbf{u}}_j(t) = \frac{1}{\sqrt{N_{xy}}} \sum_{\mathbf{k},\lambda}^{BZ} \sqrt{\frac{\hbar}{2m\omega_\lambda(\mathbf{k})}}\, \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{R}_j} (e^{-i\omega_\lambda(\mathbf{k})t} \hat{b}_{\mathbf{k}\lambda} + e^{i\omega_\lambda(-\mathbf{k})t} \hat{b}^\dagger_{-\mathbf{k}\lambda})\,, \tag{D.7}$$

N_{xy} is the number of particles in the xy-plane, $\omega_\lambda(\mathbf{k})$ and $\boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k})$ are the frequency and polarization vector for the normal mode with polarization λ, wave vector $\mathbf{k}$ and p_j indicates the layer in which the j-th atom is located.[1]

We observe that the frequencies $\omega_\lambda(\mathbf{k})$ are even under inversion $\mathbf{k} \to -\mathbf{k}$, as a result:

$$\omega_\lambda(\mathbf{k}) = \omega_\lambda(-\mathbf{k})\,. \tag{D.8}$$

Using equation (D.8), the displacement operators can be rewritten as:

$$\hat{\mathbf{u}}_j(t) = \frac{1}{\sqrt{N_{xy}}} \sum_{\mathbf{k},\lambda}^{BZ} \sqrt{\frac{\hbar}{2m\omega_\lambda(\mathbf{k})}}\, \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{R}_j} (e^{-i\omega_\lambda(\mathbf{k})t} \hat{b}_{\mathbf{k}\lambda} + e^{i\omega_\lambda(\mathbf{k})t} \hat{b}^\dagger_{-\mathbf{k}\lambda})\,. \tag{D.9}$$

[1] If $\omega_\lambda(\mathbf{k}) = 0$, this definition fails. The problem occurs for only three of the normal modes, the $\mathbf{k} = \mathbf{0}$ acoustic modes. It is a reflection of the fact that the three degrees of freedom describing translations of the entire crystal as a whole cannot be described as oscillator degrees of freedom. Only in problems in which one wishes to consider translations of the crystal as a whole, or the total momentum of the crystal, does it become important to treat these degrees of freedom correctly as well. We assume that our infinitely large harmonic crystal does not translate, so we can safely omit the $\mathbf{k} = \mathbf{0}$ modes.

The Hamiltonian for the harmonic lattice $\hat{H}_{\rm harm}$, defined in terms of phonon annihilation and creation operators, is

$$\hat{H}_{\rm harm} = \sum_{\mathbf{k},\lambda}^{BZ} \hbar\omega_\lambda(\mathbf{k})(\hat{b}^\dagger_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k}\lambda} + \frac{1}{2})\,, \tag{D.10}$$

where operators $\hat{b}_{\mathbf{k}\lambda}$ and $\hat{b}^\dagger_{\mathbf{k}\lambda}$ satisfy the usual commutation rules:

$$[\hat{b}_{\mathbf{k}\lambda}, \hat{b}_{\mathbf{k}'\lambda'}] = [\hat{b}^\dagger_{\mathbf{k}\lambda}, \hat{b}^\dagger_{\mathbf{k}'\lambda'}] = 0\,, \tag{D.11}$$

$$[\hat{b}_{\mathbf{k}\lambda}, \hat{b}^\dagger_{\mathbf{k}'\lambda'}] = \delta_2(\mathbf{k}-\mathbf{k}')\delta(\lambda-\lambda')\,. \tag{D.12}$$

As a consequence, the quantum averages between creation and annihilation operators are

$$\langle\hat{b}^\dagger_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k}'\lambda'}\rangle = \delta_2(\mathbf{k}-\mathbf{k}')\delta(\lambda-\lambda')n_\lambda(\mathbf{k})\,, \tag{D.13}$$

$$\langle\hat{b}_{\mathbf{k}\lambda}\hat{b}^\dagger_{\mathbf{k}'\lambda'}\rangle = \delta_2(\mathbf{k}-\mathbf{k}')\delta(\lambda-\lambda')(1+n_\lambda(\mathbf{k}))\,, \tag{D.14}$$

where $n_\lambda(\mathbf{k})$ is the average number of quanta in the oscillator labeled by $\mathbf{k}$ and λ at equilibrium at temperature $T = 1/(k_B\beta)$:

$$n_\lambda(\mathbf{k}) = \langle\hat{b}^\dagger_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k}\lambda}\rangle = \frac{1}{e^{\beta\hbar\omega_\lambda(\mathbf{k})}-1}\,. \tag{D.15}$$

We observe that $n_\lambda(\mathbf{k}) = n_\lambda(-\mathbf{k})$ using the parity in $\mathbf{k}$ of the phonon frequency (D.8).

A useful exercise is to calculate the commutator $[\hat{u}_{j\alpha}(t), \hat{u}_{j'\alpha'}(t')]$, where α and α' are the cartesian components x, y, z:

$$\begin{aligned}
[\hat{u}_{j\alpha}(t), \hat{u}_{j'\alpha'}(t')] &= \frac{1}{N_{xy}}\sum_{\mathbf{k},\mathbf{k}'}^{BZ}\sum_{\lambda,\lambda'}\frac{\hbar}{2m}\frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}}\epsilon_{\lambda,p_j\,\alpha}(\mathbf{k})\epsilon_{\lambda',p_{j'}\,\alpha'}(\mathbf{k}')\times \\
& e^{i\mathbf{k}\cdot\mathbf{R}_j}e^{i\mathbf{k}'\cdot\mathbf{R}_{j'}}[e^{-i\omega_\lambda(\mathbf{k})t}\hat{b}_{\mathbf{k}\lambda} + e^{i\omega_\lambda(\mathbf{k})t}\hat{b}^\dagger_{-\mathbf{k}\lambda}, e^{-i\omega_{\lambda'}(\mathbf{k}')t'}\hat{b}_{\mathbf{k}'\lambda'} + e^{i\omega_{\lambda'}(\mathbf{k}')t'}\hat{b}^\dagger_{-\mathbf{k}'\lambda'}] \\
&= \frac{1}{N_{xy}}\sum_{\mathbf{k},\mathbf{k}'}^{BZ}\sum_{\lambda,\lambda'}\frac{\hbar}{2m}\frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}}\epsilon_{\lambda,p_j\,\alpha}(\mathbf{k})\epsilon_{\lambda',p_{j'}\,\alpha'}(\mathbf{k}')e^{i(\mathbf{k}\cdot\mathbf{R}_j+\mathbf{k}'\cdot\mathbf{R}_{j'})}\times \\
& (e^{-i\omega_\lambda(\mathbf{k})t}e^{i\omega_{\lambda'}(\mathbf{k}')t'}[\hat{b}_{\mathbf{k}\lambda}, \hat{b}^\dagger_{-\mathbf{k}'\lambda'}] + e^{i\omega_\lambda(\mathbf{k})t}e^{-i\omega_{\lambda'}(\mathbf{k}')t'}[\hat{b}^\dagger_{-\mathbf{k}\lambda}, \hat{b}_{\mathbf{k}'\lambda'}]) \\
&= \frac{1}{N_{xy}}\sum_{\mathbf{k},\mathbf{k}'}^{BZ}\sum_{\lambda,\lambda'}\frac{\hbar}{2m}\frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}}\epsilon_{\lambda,p_j\,\alpha}(\mathbf{k})\epsilon_{\lambda',p_{j'}\,\alpha'}(\mathbf{k}')e^{i(\mathbf{k}\cdot\mathbf{R}_j+\mathbf{k}'\cdot\mathbf{R}_{j'})}\times \\
& \delta_2(\mathbf{k}+\mathbf{k}')\delta(\lambda-\lambda')(e^{-i\omega_\lambda(\mathbf{k})t}e^{i\omega_{\lambda'}(\mathbf{k}')t'} - e^{i\omega_\lambda(\mathbf{k})t}e^{-i\omega_{\lambda'}(\mathbf{k}')t'}) \\
&= \frac{1}{N_{xy}}\sum_{\mathbf{k},\lambda}^{BZ}\frac{\hbar}{2m\omega_\lambda(\mathbf{k})}\epsilon_{\lambda,p_j\,\alpha}(\mathbf{k})\epsilon_{\lambda,p_{j'}\,\alpha'}(-\mathbf{k})e^{i\mathbf{k}\cdot(\mathbf{R}_j-\mathbf{R}_{j'})}\times \\
& (e^{-i\omega_\lambda(\mathbf{k})(t-t')} - e^{i\omega_\lambda(\mathbf{k})(t-t')})\,.
\end{aligned} \tag{D.16}$$

It is easy to see that

$$[\hat{u}_{j\alpha}(t), \hat{u}_{j'\alpha'}(t')] = 0 \qquad \text{if} \quad t - t' = 0\,, \tag{D.17}$$

therefore, displacement operators commute if evaluated at the same time.

We start calculating the equal-time quantum averages $\langle A^2 \rangle$ and $\langle B^2 \rangle$.

$$\begin{aligned}
\langle A^2 \rangle &= \langle \Big(-i\mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t) + \xi_j \hat{u}_{jz}(t) \Big) \Big(-i\mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t) + \xi_j \hat{u}_{jz}(t) \Big) \rangle \\
&= -\langle \mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t) \mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t) \rangle - 2\,i\,\xi_j \langle \mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t) \hat{u}_{jz}(t) \rangle \\
&+ \xi_j^2 \langle \hat{u}_{jz}(t) \hat{u}_{jz}(t) \rangle \\
&= \frac{1}{N_{xy}} \sum_{\mathbf{k},\mathbf{k}'}^{BZ} \sum_{\lambda,\lambda'} \frac{\hbar}{2m} \frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}} e^{i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{R}_j} \Big[-\mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda',p_j}(\mathbf{k}') \\
&- 2\,i\,\xi_j \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \epsilon_{\lambda',p_j\,z}(\mathbf{k}') + \xi_j^2\, \epsilon_{\lambda,p_j\,z}(\mathbf{k}) \epsilon_{\lambda',p_j\,z}(\mathbf{k}') \Big] \times \\
&\langle (e^{-i\omega_\lambda(\mathbf{k})t} \hat{b}_{\mathbf{k}\lambda} + e^{i\omega_\lambda(\mathbf{k})t} \hat{b}^\dagger_{-\mathbf{k}\lambda})(e^{-i\omega_{\lambda'}(\mathbf{k}')t} \hat{b}_{\mathbf{k}'\lambda'} + e^{i\omega_{\lambda'}(\mathbf{k}')t} \hat{b}^\dagger_{-\mathbf{k}'\lambda'}) \rangle \\
&= \frac{1}{N_{xy}} \sum_{\mathbf{k},\mathbf{k}'}^{BZ} \sum_{\lambda,\lambda'} \frac{\hbar}{2m} \frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}} e^{i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{R}_j} \Big[-\mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda',p_j}(\mathbf{k}') \\
&- 2\,i\,\xi_j \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \epsilon_{\lambda',p_j\,z}(\mathbf{k}') + \xi_j^2\, \epsilon_{\lambda,p_j\,z}(\mathbf{k}) \epsilon_{\lambda',p_j\,z}(\mathbf{k}') \Big] \times \\
&(e^{-i\omega_\lambda(\mathbf{k})t} e^{i\omega_{\lambda'}(\mathbf{k}')t} \langle \hat{b}_{\mathbf{k}\lambda} \hat{b}^\dagger_{-\mathbf{k}'\lambda'} \rangle + e^{i\omega_\lambda(\mathbf{k})t} e^{-i\omega_{\lambda'}(\mathbf{k}')t} \langle \hat{b}^\dagger_{-\mathbf{k}\lambda} \hat{b}_{\mathbf{k}'\lambda'} \rangle) \\
&= \frac{1}{N_{xy}} \sum_{\mathbf{k},\mathbf{k}'}^{BZ} \sum_{\lambda,\lambda'} \frac{\hbar}{2m} \frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}} e^{i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{R}_j} \Big[-\mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda',p_j}(\mathbf{k}') \\
&- 2\,i\,\xi_j \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \epsilon_{\lambda',p_j\,z}(\mathbf{k}') + \xi_j^2\, \epsilon_{\lambda,p_j\,z}(\mathbf{k}) \epsilon_{\lambda',p_j\,z}(\mathbf{k}') \Big] \times \\
&\delta_2(\mathbf{k}+\mathbf{k}')\delta(\lambda-\lambda')(e^{-i(\omega_\lambda(\mathbf{k})-\omega_{\lambda'}(\mathbf{k}'))t}(1+n_\lambda(\mathbf{k})) \\
&+ e^{i(\omega_\lambda(\mathbf{k})-\omega_{\lambda'}(\mathbf{k}'))t} n_\lambda(\mathbf{k})) \\
&= \frac{1}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} \Big[-\mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(-\mathbf{k}) \\
&- 2\,i\,\xi_j \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \epsilon_{\lambda,p_j\,z}(-\mathbf{k}) + \xi_j^2\, \epsilon_{\lambda,p_j\,z}(\mathbf{k}) \epsilon_{\lambda,p_j\,z}(-\mathbf{k}) \Big] (1+2n_\lambda(\mathbf{k})) \\
&\equiv -2W(\mathbf{Q}_\parallel, p_j) - 4\,i\,\xi_j\, W_1(\mathbf{Q}_\parallel, p_j) + 2\,\xi_j^2\, W_2(p_j)\,,
\end{aligned} \tag{D.18}$$

where we define:

$$W(\mathbf{Q}_\parallel, p_j) \equiv \frac{1}{2N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(-\mathbf{k})(1+2n_\lambda(\mathbf{k}))\,, \tag{D.19}$$

$$W_1(\mathbf{Q}_\parallel, p_j) \equiv \frac{1}{2N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \epsilon_{\lambda,p_j\,z}(-\mathbf{k})(1+2n_\lambda(\mathbf{k}))\,, \tag{D.20}$$

$$W_2(p_j) \equiv \frac{1}{2N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} \epsilon_{\lambda,p_j\,z}(\mathbf{k}) \epsilon_{\lambda,p_j\,z}(-\mathbf{k})(1+2n_\lambda(\mathbf{k}))\,. \tag{D.21}$$

The "standard" Debye-Waller factors W are dimensionless, while the new W_1 and W_2 factors are dimensionally a length and an area, respectively.

$$\begin{aligned}
\langle B^2\rangle &= \langle \Big(i(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \hat{\mathbf{u}}_{j'\parallel}(0) + \xi'_{j'} \hat{u}_{j'z}(0) \Big) \times \\
&\Big(i(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \hat{\mathbf{u}}_{j'\parallel}(0) + \xi'_{j'} \hat{u}_{j'z}(0) \Big) \rangle \\
&= -\langle (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \hat{\mathbf{u}}_{j'\parallel}(0) (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \hat{\mathbf{u}}_{j'\parallel}(0) \rangle \\
&+ 2\,i\,\xi'_{j'} \langle (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \hat{\mathbf{u}}_{j'\parallel}(0) \hat{u}_{j'z}(0) \rangle + \xi'^2_{j'} \langle \hat{u}_{j'z}(0) \hat{u}_{j'z}(0) \rangle \\
&= \frac{1}{N_{xy}} \sum_{\mathbf{k},\mathbf{k}'}^{BZ} \sum_{\lambda,\lambda'} \frac{\hbar}{2m} \frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}} e^{i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{R}_{j'}} \Big[-(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k}) \times \\
&(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda',p_{j'}}(\mathbf{k}') + 2\,i\,\xi'_{j'} (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k}) \epsilon_{\lambda',p_{j'}\,z}(\mathbf{k}') \\
&+ \xi'^2_{j'}\, \epsilon_{\lambda,p_{j'}\,z}(\mathbf{k}) \epsilon_{\lambda',p_{j'}\,z}(\mathbf{k}') \Big] \langle (\hat{b}_{\mathbf{k}\lambda} + \hat{b}^\dagger_{-\mathbf{k}\lambda})(\hat{b}_{\mathbf{k}'\lambda'} + \hat{b}^\dagger_{-\mathbf{k}'\lambda'}) \rangle \\
&= \frac{1}{N_{xy}} \sum_{\mathbf{k},\mathbf{k}'}^{BZ} \sum_{\lambda,\lambda'} \frac{\hbar}{2m} \frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}} e^{i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{R}_{j'}} \Big[-(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k}) \times \\
&(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda',p_{j'}}(\mathbf{k}') + 2\,i\,\xi'_{j'} (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k}) \epsilon_{\lambda',p_{j'}\,z}(\mathbf{k}') \\
&+ \xi'^2_{j'}\, \epsilon_{\lambda,p_{j'}\,z}(\mathbf{k}) \epsilon_{\lambda',p_{j'}\,z}(\mathbf{k}') \Big] (\langle \hat{b}_{\mathbf{k}\lambda} \hat{b}^\dagger_{-\mathbf{k}'\lambda'} \rangle + \langle \hat{b}^\dagger_{-\mathbf{k}\lambda} \hat{b}_{\mathbf{k}'\lambda'} \rangle) \\
&= \frac{1}{N_{xy}} \sum_{\mathbf{k},\mathbf{k}'}^{BZ} \sum_{\lambda,\lambda'} \frac{\hbar}{2m} \frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}} e^{i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{R}_{j'}} \Big[-(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k}) \times \\
&(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda',p_{j'}}(\mathbf{k}') + 2\,i\,\xi'_{j'} (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k}) \epsilon_{\lambda',p_{j'}\,z}(\mathbf{k}') \\
&+ \xi'^2_{j'}\, \epsilon_{\lambda,p_{j'}\,z}(\mathbf{k}) \epsilon_{\lambda',p_{j'}\,z}(\mathbf{k}') \Big] \delta_2(\mathbf{k}+\mathbf{k}') \delta(\lambda-\lambda')(1+2n_\lambda(\mathbf{k}))
\end{aligned}$$

$$= \frac{1}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} \Big[-(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k}) (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(-\mathbf{k})$$

$$+ 2\, i\, \xi'_{j'} (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k}) \epsilon_{\lambda,p_{j'}\, z}(-\mathbf{k}) + \xi'^2_{j'}\, \epsilon_{\lambda,p_{j'}\, z}(\mathbf{k}) \epsilon_{\lambda,p_{j'}\, z}(-\mathbf{k}) \Big] \times$$

$$(1 + 2n_\lambda(\mathbf{k}))$$

$$= -2W(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'}) + 4\, i\, \xi'_{j'}\, W_1(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'}) + 2\, \xi'^2_{j'}\, W_2(p_{j'})\,. \quad \text{(D.22)}$$

Now we calculate the quantum averages $\langle AB \rangle$, $\langle BA \rangle$ that explicitly depends on time.

$$\langle AB \rangle = \langle \Big(-i\mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t) + \xi_j \hat{u}_{jz}(t) \Big) \Big(i(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \hat{\mathbf{u}}_{j'\parallel}(0) + \xi'_{j'} \hat{u}_{j'z}(0) \Big) \rangle$$

$$= \langle \mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t) (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \hat{\mathbf{u}}_{j'\parallel}(0) \rangle - i\, \xi'_{j'} \langle \mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t) \hat{u}_{j'z}(0) \rangle$$

$$+ i\, \xi_j \langle \hat{u}_{jz}(t) (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \hat{\mathbf{u}}_{j'\parallel}(0) \rangle + \xi_j\, \xi'_{j'} \langle \hat{u}_{jz}(t) \hat{u}_{j'z}(0) \rangle$$

$$= \frac{1}{N_{xy}} \sum_{\mathbf{k},\mathbf{k}'}^{BZ} \sum_{\lambda,\lambda'} \frac{\hbar}{2m} \frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}} e^{i(\mathbf{k}\cdot\mathbf{R}_j + \mathbf{k}'\cdot\mathbf{R}_{j'})} \Big[\mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \times$$

$$(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda',p_{j'}}(\mathbf{k}') - i\, \xi'_{j'}\, \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \epsilon_{\lambda',p_{j'}\, z}(\mathbf{k}')$$

$$+ i\, \xi_j\, \epsilon_{\lambda,p_j\, z}(\mathbf{k}) (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda',p_{j'}}(\mathbf{k}') + \xi_j\, \xi'_{j'}\, \epsilon_{\lambda,p_j\, z}(\mathbf{k}) \epsilon_{\lambda',p_{j'}\, z}(\mathbf{k}') \Big] \times$$

$$\langle (e^{-i\omega_\lambda(\mathbf{k})t} \hat{b}_{\mathbf{k}\lambda} + e^{i\omega_\lambda(\mathbf{k})t} \hat{b}^\dagger_{-\mathbf{k}\lambda})(\hat{b}_{\mathbf{k}'\lambda'} + \hat{b}^\dagger_{-\mathbf{k}'\lambda'}) \rangle$$

$$= \frac{1}{N_{xy}} \sum_{\mathbf{k},\mathbf{k}'}^{BZ} \sum_{\lambda,\lambda'} \frac{\hbar}{2m} \frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}} e^{i(\mathbf{k}\cdot\mathbf{R}_j + \mathbf{k}'\cdot\mathbf{R}_{j'})} \Big[\mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \times$$

$$(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda',p_{j'}}(\mathbf{k}') - i\, \xi'_{j'}\, \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \epsilon_{\lambda',p_{j'}\, z}(\mathbf{k}')$$

$$+ i\, \xi_j\, \epsilon_{\lambda,p_j\, z}(\mathbf{k}) (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda',p_{j'}}(\mathbf{k}') + \xi_j\, \xi'_{j'}\, \epsilon_{\lambda,p_j\, z}(\mathbf{k}) \epsilon_{\lambda',p_{j'}\, z}(\mathbf{k}') \Big] \times$$

$$(e^{-i\omega_\lambda(\mathbf{k})t} \langle \hat{b}_{\mathbf{k}\lambda} \hat{b}^\dagger_{-\mathbf{k}'\lambda'} \rangle + e^{i\omega_\lambda(\mathbf{k})t} \langle \hat{b}^\dagger_{-\mathbf{k}\lambda} \hat{b}_{\mathbf{k}'\lambda'} \rangle)$$

$$= \frac{1}{N_{xy}} \sum_{\mathbf{k},\mathbf{k}'}^{BZ} \sum_{\lambda,\lambda'} \frac{\hbar}{2m} \frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}} e^{i(\mathbf{k}\cdot\mathbf{R}_j + \mathbf{k}'\cdot\mathbf{R}_{j'})} \Big[\mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \times$$

$$(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda',p_{j'}}(\mathbf{k}') - i\, \xi'_{j'}\, \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \epsilon_{\lambda',p_{j'}\, z}(\mathbf{k}')$$

$$+ i\, \xi_j\, \epsilon_{\lambda,p_j\, z}(\mathbf{k}) (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda',p_{j'}}(\mathbf{k}') + \xi_j\, \xi'_{j'}\, \epsilon_{\lambda,p_j\, z}(\mathbf{k}) \epsilon_{\lambda',p_{j'}\, z}(\mathbf{k}') \Big] \times$$

$$\delta_2(\mathbf{k} + \mathbf{k}') \delta(\lambda - \lambda') [e^{-i\omega_\lambda(\mathbf{k})t} (1 + n_\lambda(\mathbf{k})) + e^{i\omega_\lambda(\mathbf{k})t} n_\lambda(\mathbf{k})]$$

$$= \frac{1}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} e^{i\mathbf{k}\cdot(\mathbf{R}_j - \mathbf{R}_{j'})} \Big[\mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k})(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(-\mathbf{k})$$
$$- i\,\xi'_{j'}\,\mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \epsilon_{\lambda,p_{j'}\,z}(-\mathbf{k}) + i\,\xi_j\,\epsilon_{\lambda,p_j\,z}(\mathbf{k})(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(-\mathbf{k})$$
$$+ \xi_j\,\xi'_{j'}\,\epsilon_{\lambda,p_j\,z}(\mathbf{k}) \epsilon_{\lambda,p_{j'}\,z}(-\mathbf{k}) \Big] [\cos(\omega_\lambda(\mathbf{k})t)(1 + 2n_\lambda(\mathbf{k})) - i\sin(\omega_\lambda(\mathbf{k})t)]$$
$$\equiv \phi_{jj'}(t,\beta) - i\,\xi'_{j'}\,\phi^{(1a)}_{jj'}(t,\beta) + i\,\xi_j\,\phi^{(1b)}_{jj'}(t,\beta) + \xi_j\,\xi'_{j'}\,\phi^{(2)}_{jj'}(t,\beta)\,, \qquad \text{(D.23)}$$

where we define:

$$\phi_{jj'}(t,\beta) \equiv \frac{1}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} e^{i\mathbf{k}\cdot(\mathbf{R}_j - \mathbf{R}_{j'})} \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \times$$
$$(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(-\mathbf{k}) [\cos(\omega_\lambda(\mathbf{k})t)(1 + 2n_\lambda(\mathbf{k})) - i\sin(\omega_\lambda(\mathbf{k})t)]\,, \qquad \text{(D.24)}$$

$$\phi^{(1a)}_{jj'}(t,\beta) \equiv \frac{1}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} e^{i\mathbf{k}\cdot(\mathbf{R}_j - \mathbf{R}_{j'})} \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \epsilon_{\lambda,p_{j'}\,z}(-\mathbf{k}) \times$$
$$[\cos(\omega_\lambda(\mathbf{k})t)(1 + 2n_\lambda(\mathbf{k})) - i\sin(\omega_\lambda(\mathbf{k})t)]\,, \qquad \text{(D.25)}$$

$$\phi^{(1b)}_{jj'}(t,\beta) \equiv \frac{1}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} e^{i\mathbf{k}\cdot(\mathbf{R}_j - \mathbf{R}_{j'})} \epsilon_{\lambda,p_j\,z}(\mathbf{k})(\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(-\mathbf{k}) \times$$
$$[\cos(\omega_\lambda(\mathbf{k})t)(1 + 2n_\lambda(\mathbf{k})) - i\sin(\omega_\lambda(\mathbf{k})t)]\,, \qquad \text{(D.26)}$$

$$\phi^{(2)}_{jj'}(t,\beta) \equiv \frac{1}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} e^{i\mathbf{k}\cdot(\mathbf{R}_j - \mathbf{R}_{j'})} \epsilon_{\lambda,p_j\,z}(\mathbf{k}) \epsilon_{\lambda,p_{j'}\,z}(-\mathbf{k}) \times$$
$$[\cos(\omega_\lambda(\mathbf{k})t)(1 + 2n_\lambda(\mathbf{k})) - i\sin(\omega_\lambda(\mathbf{k})t)]\,. \qquad \text{(D.27)}$$

The time-dependent contributions $\phi_{jj'}$ are dimensionless, while $\phi^{(1a,\,1b)}_{jj'}$ and $\phi^{(2)}_{jj'}$ are dimensionally a length and an area, respectively.

$$\langle BA\rangle = \langle\Big(i(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0) + \xi'_{j'}\hat{u}_{j'z}(0)\Big)\Big(-i\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t) + \xi_j\hat{u}_{jz}(t)\Big)\rangle$$
$$= \langle(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0)\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t)\rangle + i\,\xi_j\langle(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0)\hat{u}_{jz}(t)\rangle$$
$$-\,i\,\xi'_{j'}\langle\hat{u}_{j'z}(0)\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t)\rangle + \xi_j\,\xi'_{j'}\langle\hat{u}_{j'z}(0)\hat{u}_{jz}(t)\rangle$$
$$= \frac{1}{N_{xy}}\sum_{\mathbf{k},\mathbf{k}'}^{BZ}\sum_{\lambda,\lambda'}\frac{\hbar}{2m}\frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}}e^{i(\mathbf{k}\cdot\mathbf{R}_{j'}+\mathbf{k}'\cdot\mathbf{R}_j)}\Big[(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot\boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k})\times$$
$$\mathbf{Q}_\parallel\cdot\boldsymbol{\epsilon}_{\lambda',p_j}(\mathbf{k}') + i\,\xi_j(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot\boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k})\epsilon_{\lambda',p_j\,z}(\mathbf{k}')$$
$$-\,i\,\xi'_{j'}\,\epsilon_{\lambda,p_{j'}\,z}(\mathbf{k})\mathbf{Q}_\parallel\cdot\boldsymbol{\epsilon}_{\lambda',p_j}(\mathbf{k}') + \xi_j\,\xi'_{j'}\,\epsilon_{\lambda,p_{j'}\,z}(\mathbf{k})\epsilon_{\lambda',p_j\,z}(\mathbf{k}')\Big]\times$$
$$\langle(\hat{b}_{\mathbf{k}\lambda} + \hat{b}^\dagger_{-\mathbf{k}\lambda})(e^{-i\omega_{\lambda'}(\mathbf{k}')t}\hat{b}_{\mathbf{k}'\lambda'} + e^{i\omega_{\lambda'}(\mathbf{k}')t}\hat{b}^\dagger_{-\mathbf{k}'\lambda'})\rangle$$
$$= \frac{1}{N_{xy}}\sum_{\mathbf{k},\mathbf{k}'}^{BZ}\sum_{\lambda,\lambda'}\frac{\hbar}{2m}\frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}}e^{i(\mathbf{k}\cdot\mathbf{R}_{j'}+\mathbf{k}'\cdot\mathbf{R}_j)}\Big[(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot\boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k})\times$$
$$\mathbf{Q}_\parallel\cdot\boldsymbol{\epsilon}_{\lambda',p_j}(\mathbf{k}') + i\,\xi_j(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot\boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k})\epsilon_{\lambda',p_j\,z}(\mathbf{k}')$$
$$-\,i\,\xi'_{j'}\,\epsilon_{\lambda,p_{j'}\,z}(\mathbf{k})\mathbf{Q}_\parallel\cdot\boldsymbol{\epsilon}_{\lambda',p_j}(\mathbf{k}') + \xi_j\,\xi'_{j'}\,\epsilon_{\lambda,p_{j'}\,z}(\mathbf{k})\epsilon_{\lambda',p_j\,z}(\mathbf{k}')\Big]\times$$
$$(e^{i\omega_{\lambda'}(\mathbf{k}')t}\,\langle\hat{b}_{\mathbf{k}\lambda}\hat{b}^\dagger_{-\mathbf{k}'\lambda'}\rangle + e^{-i\omega_{\lambda'}(\mathbf{k}')t}\,\langle\hat{b}^\dagger_{-\mathbf{k}\lambda}\hat{b}_{\mathbf{k}'\lambda'}\rangle)$$
$$= \frac{1}{N_{xy}}\sum_{\mathbf{k},\mathbf{k}'}^{BZ}\sum_{\lambda,\lambda'}\frac{\hbar}{2m}\frac{1}{\sqrt{\omega_\lambda(\mathbf{k})\omega_{\lambda'}(\mathbf{k}')}}e^{i(\mathbf{k}\cdot\mathbf{R}_{j'}+\mathbf{k}'\cdot\mathbf{R}_j)}\Big[(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot\boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k})\times$$
$$\mathbf{Q}_\parallel\cdot\boldsymbol{\epsilon}_{\lambda',p_j}(\mathbf{k}') + i\,\xi_j(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot\boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k})\epsilon_{\lambda',p_j\,z}(\mathbf{k}')$$
$$-\,i\,\xi'_{j'}\,\epsilon_{\lambda,p_{j'}\,z}(\mathbf{k})\mathbf{Q}_\parallel\cdot\boldsymbol{\epsilon}_{\lambda',p_j}(\mathbf{k}') + \xi_j\,\xi'_{j'}\,\epsilon_{\lambda,p_{j'}\,z}(\mathbf{k})\epsilon_{\lambda',p_j\,z}(\mathbf{k}')\Big]\times$$
$$\delta_2(\mathbf{k}+\mathbf{k}')\delta(\lambda-\lambda')[e^{i\omega_{\lambda'}(\mathbf{k}')t}(1+n_\lambda(\mathbf{k})) + e^{-i\omega_{\lambda'}(\mathbf{k}')t}n_\lambda(\mathbf{k})]$$
$$= \frac{1}{N_{xy}}\sum_{\mathbf{k},\lambda}^{BZ}\frac{\hbar}{2m\omega_\lambda(\mathbf{k})}e^{i\mathbf{k}\cdot(\mathbf{R}_{j'}-\mathbf{R}_j)}\Big[(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot\boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k})\mathbf{Q}_\parallel\cdot\boldsymbol{\epsilon}_{\lambda,p_j}(-\mathbf{k})$$
$$+\,i\,\xi_j(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot\boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k})\epsilon_{\lambda,p_j\,z}(-\mathbf{k}) - i\,\xi'_{j'}\,\epsilon_{\lambda,p_{j'}\,z}(\mathbf{k})\mathbf{Q}_\parallel\cdot\boldsymbol{\epsilon}_{\lambda,p_j}(-\mathbf{k})$$
$$+\,\xi_j\,\xi'_{j'}\,\epsilon_{\lambda,p_{j'}\,z}(\mathbf{k})\epsilon_{\lambda,p_j\,z}(-\mathbf{k})\Big][\cos(\omega_\lambda(\mathbf{k})t)(1+2n_\lambda(\mathbf{k})) + i\sin(\omega_\lambda(\mathbf{k})t)]$$
$$\equiv \tilde{\phi}_{j'j}(t,\beta) - i\,\xi'_{j'}\,\tilde{\phi}^{(1a)}_{j'j}(t,\beta) + i\,\xi_j\,\tilde{\phi}^{(1b)}_{j'j}(t,\beta) + \xi_j\,\xi'_{j'}\,\tilde{\phi}^{(2)}_{j'j}(t,\beta)\,, \qquad \text{(D.28)}$$

where we define:

$$\tilde{\phi}_{j'j}(t,\beta) \equiv \frac{1}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} e^{i\mathbf{k}\cdot(\mathbf{R}_{j'}-\mathbf{R}_j)} (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k}) \times$$
$$\mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(-\mathbf{k}) [\cos(\omega_\lambda(\mathbf{k})t)(1+2n_\lambda(\mathbf{k})) + i\sin(\omega_\lambda(\mathbf{k})t)]\,, \quad \text{(D.29)}$$

$$\tilde{\phi}^{(1a)}_{j'j}(t,\beta) \equiv \frac{1}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} e^{i\mathbf{k}\cdot(\mathbf{R}_{j'}-\mathbf{R}_j)} \epsilon_{\lambda,p_{j'}\,z}(\mathbf{k}) \mathbf{Q}_\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(-\mathbf{k}) \times$$
$$[\cos(\omega_\lambda(\mathbf{k})t)(1+2n_\lambda(\mathbf{k})) + i\sin(\omega_\lambda(\mathbf{k})t)]\,, \quad \text{(D.30)}$$

$$\tilde{\phi}^{(1b)}_{j'j}(t,\beta) \equiv \frac{1}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} e^{i\mathbf{k}\cdot(\mathbf{R}_{j'}-\mathbf{R}_j)} (\mathbf{Q}_\parallel + \mathbf{G}_y) \cdot \boldsymbol{\epsilon}_{\lambda,p_{j'}}(\mathbf{k}) \epsilon_{\lambda,p_j\,z}(-\mathbf{k}) \times$$
$$[\cos(\omega_\lambda(\mathbf{k})t)(1+2n_\lambda(\mathbf{k})) + i\sin(\omega_\lambda(\mathbf{k})t)]\,, \quad \text{(D.31)}$$

$$\tilde{\phi}^{(2)}_{j'j}(t,\beta) \equiv \frac{1}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \frac{\hbar}{2m\omega_\lambda(\mathbf{k})} e^{i\mathbf{k}\cdot(\mathbf{R}_{j'}-\mathbf{R}_j)} \epsilon_{\lambda,p_{j'}\,z}(\mathbf{k}) \epsilon_{\lambda,p_j\,z}(-\mathbf{k}) \times$$
$$[\cos(\omega_\lambda(\mathbf{k})t)(1+2n_\lambda(\mathbf{k})) + i\sin(\omega_\lambda(\mathbf{k})t)]\,. \quad \text{(D.32)}$$

The time-dependent contributions $\tilde{\phi}_{jj'}$ are dimensionless, while $\tilde{\phi}^{(1a,\,1b)}_{jj'}$ and $\tilde{\phi}^{(2)}_{jj'}$ are dimensionally a length and an area, respectively. Now we observe that the dynamic matrix for a 3D crystal slab, as shown in Appendix F, is Hermitian. Hence, its eigenvectors, the polarization vectors, satisfy the following relation:

$$\boldsymbol{\epsilon}_{\lambda,p_j}(-\mathbf{k}) = \boldsymbol{\epsilon}^\dagger_{\lambda,p_j}(\mathbf{k})\,, \quad \text{(D.33)}$$

and therefore:

$$\tilde{\phi}_{j'j}(t,\beta) = \phi^\dagger_{jj'}(t,\beta)\,, \quad \tilde{\phi}^{(*)}_{j'j}(t,\beta) = \phi^{\dagger\,(*)}_{jj'}(t,\beta)\,. \quad \text{(D.34)}$$

Let us start calculating $A0$:

$$A0 \equiv \tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz}) \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z_0 - R_{j'z}) \times$$
$$\langle [e^{-i\mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t)}, e^{i(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot \hat{\mathbf{u}}_{j'\parallel}(0)}] \rangle$$
$$= \tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz}) \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z_0 - R_{j'z}) (J(0,0) - J'(0,0))\,. \quad \text{(D.35)}$$

Using the Gaussian identity (D.4), we can write an explicit expression for $A0$:

$$A0 = \tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz}) \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z_0 - R_{j'z}) \times$$
$$e^{-W(\mathbf{Q}_\parallel, p_j)} e^{-W(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'})} (e^{\phi_{jj'}(t,\beta)} - e^{\phi^\dagger_{jj'}(t,\beta)})\,. \quad \text{(D.36)}$$

Now we focus our attention on the first perturbative order terms $A1$ and $B1$.

$$\begin{aligned}
A1 &= \tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz}) \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z')}{\partial z'}\bigg|_{z'=z_0-R_{j'z}} \times \\
&\langle [e^{-i\mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t)}, e^{i(\mathbf{Q}_\parallel+\mathbf{G}_y)\cdot \hat{\mathbf{u}}_{j'\parallel}(0)} \hat{u}_{j'z}(0)]\rangle \\
&= \tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz}) \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z')}{\partial z'}\bigg|_{z'=z_0-R_{j'z}} \times \\
&\left(\frac{\partial}{\partial \xi'_{j'}} J(\xi_j, \xi'_{j'})\Big|_{\xi_j,\xi'_{j'}=0} - \frac{\partial}{\partial \xi'_{j'}} J'(\xi_j, \xi'_{j'})\Big|_{\xi_j,\xi'_{j'}=0} \right) \\
&= \tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz}) \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z')}{\partial z'}\bigg|_{z'=z_0-R_{j'z}} \times \\
&\bigg[e^{-W(\mathbf{Q}_\parallel, p_j)} e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y, p_{j'})} e^{\phi_{jj'}(t,\beta)} \bigg(2\, i\, W_1(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'}) \\
&- i\, \phi^{(1a)}_{jj'}(t,\beta) \bigg) - e^{-W(\mathbf{Q}_\parallel, p_j)} e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y, p_{j'})} e^{\phi^\dagger_{jj'}(t,\beta)} \times \\
&\bigg(2\, i\, W_1(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'}) - i\, \phi^{\dagger(1a)}_{jj'}(t,\beta) \bigg) \bigg] \\
&= \tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz}) \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z')}{\partial z'}\bigg|_{z'=z_0-R_{j'z}} \times \\
&e^{-W(\mathbf{Q}_\parallel, p_j)} e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y, p_{j'})} \bigg[e^{\phi_{jj'}(t,\beta)} \bigg(2\, i\, W_1(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'}) \\
&- i\, \phi^{(1a)}_{jj'}(t,\beta) \bigg) - e^{\phi^\dagger_{jj'}(t,\beta)} \bigg(2\, i\, W_1(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'}) - i\, \phi^{\dagger(1a)}_{jj'}(t,\beta) \bigg) \bigg].
\end{aligned} \tag{D.37}$$

$$
\begin{aligned}
B1 = & \left.\frac{\partial\tilde{V}(|\mathbf{Q}_\parallel|, z)}{\partial z}\right|_{z=z_0-R_{jz}} \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z_0 - R_{j'z})\times \\
& \langle[e^{-i\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t)}\hat{u}_{jz}(t), e^{i(\mathbf{Q}_\parallel+\mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0)}]\rangle \\
= & \left.\frac{\partial\tilde{V}(|\mathbf{Q}_\parallel|, z)}{\partial z}\right|_{z=z_0-R_{jz}} \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z_0 - R_{j'z})\times \\
& \left(\frac{\partial}{\partial\xi_j}J(\xi_j,\xi'_{j'})\Big|_{\xi_j,\xi'_{j'}=0} - \frac{\partial}{\partial\xi_j}J'(\xi_j,\xi'_{j'})\Big|_{\xi_j,\xi'_{j'}=0}\right) \\
= & \left.\frac{\partial\tilde{V}(|\mathbf{Q}_\parallel|, z)}{\partial z}\right|_{z=z_0-R_{jz}} \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z_0 - R_{j'z})\times \\
& \Big[e^{-W(\mathbf{Q}_\parallel,p_j)}e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'})}e^{\phi_{jj'}(t,\beta)}\left(-2\,i\,W_1(\mathbf{Q}_\parallel,p_j) + i\,\phi^{(1b)}_{jj'}(t,\beta)\right) \\
& - e^{-W(\mathbf{Q}_\parallel,p_j)}e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'})}e^{\phi^\dagger_{jj'}(t,\beta)}\left(-2\,i\,W_1(\mathbf{Q}_\parallel,p_j) + i\,\phi^{\dagger(1b)}_{jj'}(t,\beta)\right)\Big] \\
= & \left.\frac{\partial\tilde{V}(|\mathbf{Q}_\parallel|, z)}{\partial z}\right|_{z=z_0-R_{jz}} \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z_0 - R_{j'z})\times \\
& e^{-W(\mathbf{Q}_\parallel,p_j)}e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'})}\Big[e^{\phi_{jj'}(t,\beta)}\left(-2\,i\,W_1(\mathbf{Q}_\parallel,p_j) + i\,\phi^{(1b)}_{jj'}(t,\beta)\right) \\
& - e^{\phi^\dagger_{jj'}(t,\beta)}\left(-2\,i\,W_1(\mathbf{Q}_\parallel,p_j) + i\,\phi^{\dagger(1b)}_{jj'}(t,\beta)\right)\Big]\,. \qquad \text{(D.38)}
\end{aligned}
$$

Finally we can calculate the second perturbative order terms $A2$, $B2$ and $C2$.

$$
\begin{aligned}
A2 = & \frac{1}{2}\tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz})\left.\frac{\partial^2\tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z')}{\partial z'^2}\right|_{z'=z_0-R_{j'z}}\times \\
& \langle[e^{-i\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t)}, e^{i(\mathbf{Q}_\parallel+\mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0)}(\hat{u}_{j'z}(0))^2]\rangle \\
= & \frac{1}{2}\tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz})\left.\frac{\partial^2\tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z')}{\partial z'^2}\right|_{z'=z_0-R_{j'z}}\times \\
& \left(\frac{\partial^2}{\partial\xi'^2_{j'}}J(\xi_j,\xi'_{j'})\Big|_{\xi_j,\xi'_{j'}=0} - \frac{\partial^2}{\partial\xi'^2_{j'}}J'(\xi_j,\xi'_{j'})\Big|_{\xi_j,\xi'_{j'}=0}\right)
\end{aligned}
$$

$$
\begin{aligned}
&= \frac{1}{2}\tilde{V}(|\mathbf{Q}_{\parallel}|, z_0 - R_{jz}) \frac{\partial^2 \tilde{V}(|\mathbf{Q}_{\parallel} + \mathbf{G}_y|, z')}{\partial z'^2}\bigg|_{z'=z_0-R_{j'z}} \times \\
&\bigg\{ e^{-W(\mathbf{Q}_{\parallel},p_j)} e^{-W(\mathbf{Q}_{\parallel}+\mathbf{G}_y,p_{j'})} e^{\phi_{jj'}(t,\beta)} \bigg[\bigg(2\, i\, W_1(\mathbf{Q}_{\parallel} + \mathbf{G}_y, p_{j'}) \\
&- i\,\phi^{(1a)}_{jj'}(t,\beta)\bigg)^2 + 2W_2(p_{j'})\bigg] - e^{-W(\mathbf{Q}_{\parallel},p_j)} e^{-W(\mathbf{Q}_{\parallel}+\mathbf{G}_y,p_{j'})} e^{\phi^{\dagger}_{jj'}(t,\beta)} \times \\
&\bigg[\bigg(2\, i\, W_1(\mathbf{Q}_{\parallel} + \mathbf{G}_y, p_{j'}) - i\,\phi^{\dagger(1a)}_{jj'}(t,\beta)\bigg)^2 + 2W_2(p_{j'})\bigg]\bigg\} \\
&= \frac{1}{2}\tilde{V}(|\mathbf{Q}_{\parallel}|, z_0 - R_{jz}) \frac{\partial^2 \tilde{V}(|\mathbf{Q}_{\parallel} + \mathbf{G}_y|, z')}{\partial z'^2}\bigg|_{z'=z_0-R_{j'z}} \times \\
&e^{-W(\mathbf{Q}_{\parallel},p_j)} e^{-W(\mathbf{Q}_{\parallel}+\mathbf{G}_y,p_{j'})} \bigg\{ e^{\phi_{jj'}(t,\beta)} \bigg[\bigg(2\, i\, W_1(\mathbf{Q}_{\parallel} + \mathbf{G}_y, p_{j'}) \\
&- i\,\phi^{(1a)}_{jj'}(t,\beta)\bigg)^2 + 2W_2(p_{j'})\bigg] - e^{\phi^{\dagger}_{jj'}(t,\beta)} \bigg[\bigg(2\, i\, W_1(\mathbf{Q}_{\parallel} + \mathbf{G}_y, p_{j'}) \\
&- i\,\phi^{\dagger(1a)}_{jj'}(t,\beta)\bigg)^2 + 2W_2(p_{j'})\bigg]\bigg\} \\
&= \frac{1}{2}\tilde{V}(|\mathbf{Q}_{\parallel}|, z_0 - R_{jz}) \frac{\partial^2 \tilde{V}(|\mathbf{Q}_{\parallel} + \mathbf{G}_y|, z')}{\partial z'^2}\bigg|_{z'=z_0-R_{j'z}} \times \\
&e^{-W(\mathbf{Q}_{\parallel},p_j)} e^{-W(\mathbf{Q}_{\parallel}+\mathbf{G}_y,p_{j'})} \bigg\{ e^{\phi_{jj'}(t,\beta)} \bigg[-\bigg(2W_1(\mathbf{Q}_{\parallel} + \mathbf{G}_y, p_{j'}) - \phi^{(1a)}_{jj'}(t,\beta)\bigg)^2 \\
&+ 2W_2(p_{j'})\bigg] - e^{\phi^{\dagger}_{jj'}(t,\beta)} \bigg[-\bigg(2W_1(\mathbf{Q}_{\parallel} + \mathbf{G}_y, p_{j'}) - \phi^{\dagger(1a)}_{jj'}(t,\beta)\bigg)^2 \\
&+ 2W_2(p_{j'})\bigg]\bigg\}. \qquad \text{(D.39)}
\end{aligned}
$$

$$
\begin{aligned}
B2 = &\frac{1}{2} \frac{\partial^2 \tilde{V}(|\mathbf{Q}_{\parallel}|, z)}{\partial z^2}\bigg|_{z=z_0-R_{jz}} \tilde{V}(|\mathbf{Q}_{\parallel} + \mathbf{G}_y|, z_0 - R_{j'z}) \times \\
&\langle [e^{-i\mathbf{Q}_{\parallel}\cdot \hat{\mathbf{u}}_{j\parallel}(t)} (\hat{u}_{jz}(t))^2, e^{i(\mathbf{Q}_{\parallel}+\mathbf{G}_y)\cdot \hat{\mathbf{u}}_{j'\parallel}(0)}] \rangle \\
&= \frac{1}{2} \frac{\partial^2 \tilde{V}(|\mathbf{Q}_{\parallel}|, z)}{\partial z^2}\bigg|_{z=z_0-R_{jz}} \tilde{V}(|\mathbf{Q}_{\parallel} + \mathbf{G}_y|, z_0 - R_{j'z}) \times \\
&\bigg(\frac{\partial^2}{\partial \xi_j^2} J(\xi_j, \xi'_{j'})\bigg|_{\xi_j,\xi'_{j'}=0} - \frac{\partial^2}{\partial \xi_j^2} J'(\xi_j, \xi'_{j'})\bigg|_{\xi_j,\xi'_{j'}=0} \bigg)
\end{aligned}
$$

$$
\begin{aligned}
&= \frac{1}{2}\frac{\partial^2 \tilde{V}(|\mathbf{Q}_\parallel|,z)}{\partial z^2}\bigg|_{z=z_0-R_{jz}} \tilde{V}(|\mathbf{Q}_\parallel+\mathbf{G}_y|,z_0-R_{j'z})\times \\
&\bigg\{e^{-W(\mathbf{Q}_\parallel,p_j)}e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'})}e^{\phi_{jj'}(t,\beta)}\bigg[\Big(-2\,i\,W_1(\mathbf{Q}_\parallel,p_j)+i\,\phi^{(1b)}_{jj'}(t,\beta)\Big)^2 \\
&+2W_2(p_j)\bigg] - e^{-W(\mathbf{Q}_\parallel,p_j)}e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'})}e^{\phi^\dagger_{jj'}(t,\beta)}\bigg[\Big(-2\,i\,W_1(\mathbf{Q}_\parallel,p_j) \\
&+i\,\phi^{\dagger(1b)}_{jj'}(t,\beta)\Big)^2+2W_2(p_j)\bigg]\bigg\} \\
&= \frac{1}{2}\frac{\partial^2 \tilde{V}(|\mathbf{Q}_\parallel|,z)}{\partial z^2}\bigg|_{z=z_0-R_{jz}} \tilde{V}(|\mathbf{Q}_\parallel+\mathbf{G}_y|,z_0-R_{j'z})\times \\
&e^{-W(\mathbf{Q}_\parallel,p_j)}e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'})}\bigg\{e^{\phi_{jj'}(t,\beta)}\bigg[\Big(-2\,i\,W_1(\mathbf{Q}_\parallel,p_j)+i\,\phi^{(1b)}_{jj'}(t,\beta)\Big)^2 \\
&+2W_2(p_j)\bigg] - e^{\phi^\dagger_{jj'}(t,\beta)}\bigg[\Big(-2\,i\,W_1(\mathbf{Q}_\parallel,p_j)+i\,\phi^{\dagger(1b)}_{jj'}(t,\beta)\Big)^2+2W_2(p_j)\bigg]\bigg\} \\
&= \frac{1}{2}\frac{\partial^2 \tilde{V}(|\mathbf{Q}_\parallel|,z)}{\partial z^2}\bigg|_{z=z_0-R_{jz}} \tilde{V}(|\mathbf{Q}_\parallel+\mathbf{G}_y|,z_0-R_{j'z})\times \\
&e^{-W(\mathbf{Q}_\parallel,p_j)}e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'})}\bigg\{e^{\phi_{jj'}(t,\beta)}\bigg[-\Big(-2W_1(\mathbf{Q}_\parallel,p_j)+\phi^{(1b)}_{jj'}(t,\beta)\Big)^2 \\
&+2W_2(p_j)\bigg] - e^{\phi^\dagger_{jj'}(t,\beta)}\bigg[-\Big(-2W_1(\mathbf{Q}_\parallel,p_j)+\phi^{\dagger(1b)}_{jj'}(t,\beta)\Big)^2+2W_2(p_j)\bigg]\bigg\}.
\end{aligned}
\tag{D.40}
$$

$$
\begin{aligned}
C2 &= \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel|,z)}{\partial z}\bigg|_{z=z_0-R_{jz}} \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel+\mathbf{G}_y|,z')}{\partial z'}\bigg|_{z'=z_0-R_{j'z}}\times \\
&\langle[e^{-i\mathbf{Q}_\parallel\cdot\hat{\mathbf{u}}_{j\parallel}(t)}\hat{u}_{jz}(t), e^{i(\mathbf{Q}_\parallel+\mathbf{G}_y)\cdot\hat{\mathbf{u}}_{j'\parallel}(0)}\hat{u}_{j'z}(0)]\rangle \\
&= \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel|,z)}{\partial z}\bigg|_{z=z_0-R_{jz}} \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel+\mathbf{G}_y|,z')}{\partial z'}\bigg|_{z'=z_0-R_{j'z}}\times \\
&\left(\frac{\partial^2}{\partial\xi_j\partial\xi'_{j'}}J(\xi_j,\xi'_{j'})\Big|_{\xi_j,\xi'_{j'}=0} - \frac{\partial^2}{\partial\xi_j\partial\xi'_{j'}}J'(\xi_j,\xi'_{j'})\Big|_{\xi_j,\xi'_{j'}=0}\right)
\end{aligned}
$$

$$= \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel|, z)}{\partial z}\Big|_{z=z_0-R_{jz}} \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z')}{\partial z'}\Big|_{z'=z_0-R_{j'z}} \times$$
$$\Big\{ e^{-W(\mathbf{Q}_\parallel, p_j)} e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y, p_{j'})} e^{\phi_{jj'}(t,\beta)} \Big[\Big(-2\, i\, W_1(\mathbf{Q}_\parallel, p_j) + i\, \phi^{(1b)}_{jj'}(t,\beta) \Big) \times$$
$$\Big(2\, i\, W_1(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'}) - i\, \phi^{(1a)}_{jj'}(t,\beta) \Big) + \phi^{(2)}_{jj'}(t,\beta) \Big] - e^{-W(\mathbf{Q}_\parallel, p_j)} \times$$
$$e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y, p_{j'})} e^{\phi^{\dagger}_{jj'}(t,\beta)} \Big[\Big(-2\, i\, W_1(\mathbf{Q}_\parallel, p_j) + i\, \phi^{\dagger(1b)}_{jj'}(t,\beta) \Big) \times$$
$$\Big(2\, i\, W_1(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'}) - i\, \phi^{\dagger(1a)}_{jj'}(t,\beta) \Big) + \phi^{\dagger(2)}_{jj'}(t,\beta) \Big] \Big\}$$
$$= \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel|, z)}{\partial z}\Big|_{z=z_0-R_{jz}} \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z')}{\partial z'}\Big|_{z'=z_0-R_{j'z}} \times$$
$$e^{-W(\mathbf{Q}_\parallel, p_j)} e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y, p_{j'})} \Big\{ e^{\phi_{jj'}(t,\beta)} \Big[\Big(-2\, i\, W_1(\mathbf{Q}_\parallel, p_j) + i\, \phi^{(1b)}_{jj'}(t,\beta) \Big) \times$$
$$\Big(2\, i\, W_1(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'}) - i\, \phi^{(1a)}_{jj'}(t,\beta) \Big) + \phi^{(2)}_{jj'}(t,\beta) \Big] - e^{\phi^{\dagger}_{jj'}(t,\beta)} \times$$
$$\Big[\Big(-2\, i\, W_1(\mathbf{Q}_\parallel, p_j) + i\, \phi^{\dagger(1b)}_{jj'}(t,\beta) \Big) \Big(2\, i\, W_1(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'}) - i\, \phi^{\dagger(1a)}_{jj'}(t,\beta) \Big)$$
$$+ \phi^{\dagger(2)}_{jj'}(t,\beta) \Big] \Big\}$$
$$= \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel|, z)}{\partial z}\Big|_{z=z_0-R_{jz}} \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z')}{\partial z'}\Big|_{z'=z_0-R_{j'z}} \times$$
$$e^{-W(\mathbf{Q}_\parallel, p_j)} e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y, p_{j'})} \Big\{ e^{\phi_{jj'}(t,\beta)} \Big[- \Big(-2W_1(\mathbf{Q}_\parallel, p_j) + \phi^{(1b)}_{jj'}(t,\beta) \Big) \times$$
$$\Big(2W_1(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'}) - \phi^{(1a)}_{jj'}(t,\beta) \Big) + \phi^{(2)}_{jj'}(t,\beta) \Big] - e^{\phi^{\dagger}_{jj'}(t,\beta)} \times$$
$$\Big[- \Big(-2W_1(\mathbf{Q}_\parallel, p_j) + \phi^{\dagger(1b)}_{jj'}(t,\beta) \Big) \Big(2W_1(\mathbf{Q}_\parallel + \mathbf{G}_y, p_{j'}) - \phi^{\dagger(1a)}_{jj'}(t,\beta) \Big)$$
$$+ \phi^{\dagger(2)}_{jj'}(t,\beta) \Big] \Big\} . \tag{D.41}$$

Now we have all the elements to write the final result for the thermal average (D.1):

$$\langle [e^{-i\mathbf{Q}_\parallel \cdot \hat{\mathbf{u}}_{j\parallel}(t)} \tilde{V}(|\mathbf{Q}_\parallel|, z_0 - R_{jz} - \hat{u}_{jz}(t)), e^{i(\mathbf{Q}_\parallel + \mathbf{G}_y)\cdot \hat{\mathbf{u}}_{j'\parallel}(0)} \times$$
$$\tilde{V}(|\mathbf{Q}_\parallel + \mathbf{G}_y|, z_0 - R_{j'z} - \hat{u}_{j'z}(0))] \rangle$$

$$
\begin{aligned}
&= e^{-W(\mathbf{Q}_\parallel,p_j)} e^{-W(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'})}\Bigg\{ \tilde{V}(|\mathbf{Q}_\parallel|,z_0-R_{jz})\tilde{V}(|\mathbf{Q}_\parallel+\mathbf{G}_y|,z_0-R_{j'z})\times \\
&(e^{\phi_{jj'}(t,\beta)} - e^{\phi^\dagger_{jj'}(t,\beta)}) - \tilde{V}(|\mathbf{Q}_\parallel|,z_0-R_{jz}) \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel+\mathbf{G}_y|,z')}{\partial z'}\bigg|_{z'=z_0-R_{j'z}} \times \\
&\bigg[e^{\phi_{jj'}(t,\beta)}\Big(2\,i\,W_1(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'}) - i\,\phi^{(1a)}_{jj'}(t,\beta)\Big) - e^{\phi^\dagger_{jj'}(t,\beta)}\times \\
&\Big(2\,i\,W_1(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'}) - i\,\phi^{\dagger(1a)}_{jj'}(t,\beta)\Big)\bigg] - \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel|,z)}{\partial z}\bigg|_{z=z_0-R_{jz}} \times \\
&\tilde{V}(|\mathbf{Q}_\parallel+\mathbf{G}_y|,z_0-R_{j'z})\bigg[e^{\phi_{jj'}(t,\beta)}\Big(-2\,i\,W_1(\mathbf{Q}_\parallel,p_j) + i\,\phi^{(1b)}_{jj'}(t,\beta)\Big) \\
&- e^{\phi^\dagger_{jj'}(t,\beta)}\Big(-2\,i\,W_1(\mathbf{Q}_\parallel,p_j) + i\,\phi^{\dagger(1b)}_{jj'}(t,\beta)\Big)\bigg] + \frac{1}{2}\tilde{V}(|\mathbf{Q}_\parallel|,z_0-R_{jz})\times \\
&\frac{\partial^2 \tilde{V}(|\mathbf{Q}_\parallel+\mathbf{G}_y|,z')}{\partial z'^2}\bigg|_{z'=z_0-R_{j'z}} \bigg\{ e^{\phi_{jj'}(t,\beta)}\bigg[-\Big(2W_1(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'}) \\
&- \phi^{(1a)}_{jj'}(t,\beta)\Big)^2 + 2W_2(p_{j'})\bigg] - e^{\phi^\dagger_{jj'}(t,\beta)}\bigg[-\Big(2W_1(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'}) \\
&- \phi^{\dagger(1a)}_{jj'}(t,\beta)\Big)^2 + 2W_2(p_{j'})\bigg]\bigg\} + \frac{1}{2}\frac{\partial^2 \tilde{V}(|\mathbf{Q}_\parallel|,z)}{\partial z^2}\bigg|_{z=z_0-R_{jz}} \times \\
&\tilde{V}(|\mathbf{Q}_\parallel+\mathbf{G}_y|,z_0-R_{j'z})\bigg\{ e^{\phi_{jj'}(t,\beta)}\bigg[-\Big(-2W_1(\mathbf{Q}_\parallel,p_j) + \phi^{(1b)}_{jj'}(t,\beta)\Big)^2 \\
&+ 2W_2(p_j)\bigg] - e^{\phi^\dagger_{jj'}(t,\beta)}\bigg[-\Big(-2W_1(\mathbf{Q}_\parallel,p_j) + \phi^{\dagger(1b)}_{jj'}(t,\beta)\Big)^2 + 2W_2(p_j)\bigg]\bigg\} \\
&+ \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel|,z)}{\partial z}\bigg|_{z=z_0-R_{jz}} \frac{\partial \tilde{V}(|\mathbf{Q}_\parallel+\mathbf{G}_y|,z')}{\partial z'}\bigg|_{z'=z_0-R_{j'z}} \times \\
&\bigg\{ e^{\phi_{jj'}(t,\beta)}\bigg[-\Big(-2W_1(\mathbf{Q}_\parallel,p_j) + \phi^{(1b)}_{jj'}(t,\beta)\Big)\times \\
&\Big(2W_1(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'}) - \phi^{(1a)}_{jj'}(t,\beta)\Big) + \phi^{(2)}_{jj'}(t,\beta)\bigg] - e^{\phi^\dagger_{jj'}(t,\beta)}\times \\
&\bigg[-\Big(-2W_1(\mathbf{Q}_\parallel,p_j) + \phi^{\dagger(1b)}_{jj'}(t,\beta)\Big)\Big(2W_1(\mathbf{Q}_\parallel+\mathbf{G}_y,p_{j'}) - \phi^{\dagger(1a)}_{jj'}(t,\beta)\Big) \\
&+ \phi^{\dagger(2)}_{jj'}(t,\beta)\bigg]\bigg\}\Bigg\} + o(\hat{u}_z^2)\,. \qquad \text{(D.42)}
\end{aligned}
$$

This final expression for the thermal average is then used in Sec. 4.3 for the calculation of friction.

Appendix E

One-phonon approximation

In this Appendix we want to provide the detailed calculation of the friction by means the one-phonon approximation. For convenience, we rewrite Eq. (4.45):

$$\begin{aligned}
F = & \frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{\mathbf{G}^y} e^{-iG^y y_0} \int \frac{d^2 Q^{\parallel}}{(2\pi)^2} \mathbf{Q}^{\parallel} \cdot \hat{\mathbf{v}}_{\mathrm{SL}} \times \\
& \mathrm{Re}\Bigg(\sum_{j,p_{j'}} e^{-i\mathbf{Q}^{\parallel}\cdot\mathbf{R}_j^{\parallel}} e^{-W(\mathbf{Q}^{\parallel},p_j)} e^{-W(\mathbf{Q}^{\parallel}+\mathbf{G}^y,p_{j'})} \times \\
& \int_0^{\infty} dt\, e^{(i\mathbf{Q}^{\parallel}\cdot\mathbf{v}_{\mathrm{SL}}-\gamma/2)t} \Big(L^{(0)}_{jp_{j'}} + L^{(1)}_{jp_{j'}} + L^{(2)}_{jp_{j'}} \Big) \Bigg) .
\end{aligned} \tag{E.1}$$

We adopt the one-phonon approximation for the terms involving $\phi_{jp_{j'}}(t,\beta)$:

$$e^{\phi_{jp_{j'}}(t,\beta)} \simeq 1 + \phi_{jp_{j'}}(t,\beta)\,, \tag{E.2}$$

which is valid in the limit of small $|\phi_{jp_{j'}}(t,\beta)|$, appropriate for not too large wave vector $\mathbf{Q}^{\parallel}$ and not too high temperature.

We start to apply the one-phonon approximation to the first term of Eq. (E.1):

$$\begin{aligned}
& \frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_{\parallel}} e^{-i\mathbf{Q}^{\parallel}\cdot\mathbf{R}_j^{\parallel}} L^{(0)}_{jp_{j'}} \\
& = \frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_{\parallel}} e^{-i\mathbf{Q}^{\parallel}\cdot\mathbf{R}_j^{\parallel}} \tilde{V}(|\mathbf{Q}^{\parallel}|, z_0 - R^z_{p_j}) \tilde{V}(|\mathbf{Q}^{\parallel}+\mathbf{G}^y|, z_0 - R^z_{p_{j'}}) \times \\
& \frac{1}{\hbar}\left(e^{\phi_{jp_{j'}}(t,\beta)} - e^{\phi^{\dagger}_{jp_{j'}}(t,\beta)}\right)
\end{aligned}$$

$$= \frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel\cdot\mathbf{R}_j^\parallel} \tilde{V}(|\mathbf{Q}^\parallel|, z_0 - R^z_{p_j})\tilde{V}(|\mathbf{Q}^\parallel + \mathbf{G}^y|, z_0 - R^z_{p_{j'}})\times$$
$$\frac{1}{\hbar}(\phi_{jp_{j'}}(t,\beta) - \phi^\dagger_{jp_{j'}}(t,\beta))$$
$$= \frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel\cdot\mathbf{R}_j^\parallel} \tilde{V}(|\mathbf{Q}^\parallel|, z_0 - R^z_{p_j})\tilde{V}(|\mathbf{Q}^\parallel + \mathbf{G}^y|, z_0 - R^z_{p_{j'}})\times$$
$$\frac{1}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \Big[e^{i\mathbf{k}\cdot\mathbf{R}_j^\parallel} \Phi_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{k}, \lambda, t, \beta)$$
$$- e^{-i\mathbf{k}\cdot\mathbf{R}_j^\parallel} \Phi^\dagger_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{k}, \lambda, t, \beta) \Big], \quad \text{(E.3)}$$

where we define:

$$\Phi_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{k}, \lambda, t, \beta) \equiv \frac{1}{2m\omega_\lambda(\mathbf{k})} \mathbf{Q}^\parallel \cdot \boldsymbol{\epsilon}_{\lambda, p_j}(\mathbf{k})\times$$
$$(\mathbf{Q}^\parallel + \mathbf{G}^y)\cdot \boldsymbol{\epsilon}_{\lambda, p_{j'}}(-\mathbf{k})[\cos(\omega_\lambda(\mathbf{k})t)(1 + 2n_\lambda(\mathbf{k})) - i\sin(\omega_\lambda(\mathbf{k})t)]\,. \quad \text{(E.4)}$$

$\Phi_{p_j p_{j'}}$ are dimensionally an (Energy·Time)$^{-1}$.

Executing the thermodynamic limit

$$\lim_{N_{xy}\to\infty} \frac{1}{a^2\, N_{xy}} \sum_{\mathbf{k}}^{BZ} \longrightarrow \int_{BZ} \frac{d^2k}{(2\pi)^2}$$

we obtain:

$$\frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel\cdot\mathbf{R}_j^\parallel} L^{(0)}_{jp_{j'}}$$
$$= \sum_{j_\parallel} \tilde{V}(|\mathbf{Q}^\parallel|, z_0 - R^z_{p_j})\tilde{V}(|\mathbf{Q}^\parallel + \mathbf{G}^y|, z_0 - R^z_{p_{j'}})\times$$
$$\sum_\lambda \int_{BZ} \frac{d^2k}{(2\pi)^2} \Big[e^{-i\mathbf{R}_j^\parallel\cdot(\mathbf{Q}^\parallel - \mathbf{k})} \Phi_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{k}, \lambda, t, \beta)$$
$$- e^{-i\mathbf{R}_j^\parallel\cdot(\mathbf{Q}^\parallel + \mathbf{k})} \Phi^\dagger_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{k}, \lambda, t, \beta) \Big]. \quad \text{(E.5)}$$

Now we make use of the 2D version of the periodic delta function, or *Dirac comb*, identity:

$$\sum_{j_\parallel} e^{-i\mathbf{R}_j^\parallel\cdot(\mathbf{Q}^\parallel - \mathbf{k})} = \frac{(2\pi)^2}{a^2} \sum_{\mathbf{G}'} \delta_2(\mathbf{k} - \mathbf{Q}^\parallel - \mathbf{G}')\,, \quad \text{(E.6)}$$

$$\sum_{j_\parallel} e^{-i\mathbf{R}_j^\parallel\cdot(\mathbf{Q}^\parallel + \mathbf{k})} = \frac{(2\pi)^2}{a^2} \sum_{\mathbf{G}'} \delta_2(\mathbf{k} + \mathbf{Q}^\parallel + \mathbf{G}')\,, \quad \text{(E.7)}$$

where $\mathbf{G}' = \frac{2\pi}{a}(l^x, l^y)$ are the reciprocal lattice vectors of the 2D lattice. Hence, combining the integral over $\mathbf{k}$ extended over the first BZ with the $\mathbf{G}'$ summation, we obtain an unrestricted integral on a variable $\mathbf{k}' = \mathbf{k} - \mathbf{G}'$ for the first term of Eq. (E.5) and an unrestricted integral on a variable $\mathbf{k}' = \mathbf{k} + \mathbf{G}'$ for the second term of the same equation. Furthermore, the $\delta_2(\mathbf{k}' - \mathbf{Q}^{\parallel})$ and $\delta_2(\mathbf{k}' + \mathbf{Q}^{\parallel})$ conditions eliminate the integration over $\mathbf{k}'$ and they fix $\mathbf{k}'$ to $\mathbf{Q}^{\parallel}$ and to $-\mathbf{Q}^{\parallel}$ respectively. We rewrite Eq. (E.5) as

$$
\begin{aligned}
&\frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_\parallel} e^{-i\mathbf{Q}^{\parallel}\cdot\mathbf{R}_j^{\parallel}} L^{(0)}_{jp_{j'}} \\
&= \frac{1}{a^2} \tilde{V}(|\mathbf{Q}^{\parallel}|, z_0 - R^z_{p_j}) \tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z_0 - R^z_{p_{j'}}) \times \\
&\sum_{\lambda} \Big[\Phi_{p_j p_{j'}}(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\parallel} + \mathbf{G}^y, \mathbf{Q}^{\parallel}, \lambda, t, \beta) - \Phi^{\dagger}_{p_j p_{j'}}(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\parallel} + \mathbf{G}^y, -\mathbf{Q}^{\parallel}, \lambda, t, \beta) \Big] \\
&\equiv \mathscr{L}^{(0)}_{p_j p_{j'}}(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\parallel} + \mathbf{G}^y, t, z_0, \beta)\,. \qquad \text{(E.8)}
\end{aligned}
$$

We observe that in this low-temperature gentle-interaction regime, Planck's constant $\hbar$ disappears from the friction term based on $\mathscr{L}^{(0)}_{p_j p_{j'}}$.

We use the same technique to calculate the second term of Eq. (E.1). Applying the one-phonon approximation, we obtain:

$$
\begin{aligned}
&\frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_\parallel} e^{-i\mathbf{Q}^{\parallel}\cdot\mathbf{R}_j^{\parallel}} L^{(A1)}_{jp_{j'}} \\
&= -\frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_\parallel} e^{-i\mathbf{Q}^{\parallel}\cdot\mathbf{R}_j^{\parallel}} \tilde{V}(|\mathbf{Q}^{\parallel}|, z_0 - R^z_{p_j}) \left. \frac{\partial \tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z')}{\partial z'} \right|_{z' = z_0 - R^z_{p_{j'}}} \times \\
&\frac{1}{\hbar} \Big[e^{\phi_{jp_{j'}}(t,\beta)} \Big(2\,i\,W_1(\mathbf{Q}^{\parallel} + \mathbf{G}^y, p_{j'}) - i\,\phi^{(1a)}_{jp_{j'}}(t,\beta) \Big) \\
&\qquad - e^{\phi^{\dagger}_{jp_{j'}}(t,\beta)} \Big(2\,i\,W_1(\mathbf{Q}^{\parallel} + \mathbf{G}^y, p_{j'}) - i\,\phi^{\dagger(1a)}_{jp_{j'}}(t,\beta) \Big) \Big] \\
&= -\frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_\parallel} e^{-i\mathbf{Q}^{\parallel}\cdot\mathbf{R}_j^{\parallel}} \tilde{V}(|\mathbf{Q}^{\parallel}|, z_0 - R^z_{p_j}) \left. \frac{\partial \tilde{V}(|\mathbf{Q}^{\parallel} + \mathbf{G}^y|, z')}{\partial z'} \right|_{z' = z_0 - R^z_{p_{j'}}} \times \\
&\Big[\frac{2\,i\,W_1(\mathbf{Q}^{\parallel} + \mathbf{G}^y, p_{j'})}{\hbar} \Big(\phi_{jp_{j'}}(t,\beta) - \phi^{\dagger}_{jp_{j'}}(t,\beta) \Big) \\
&\qquad - \frac{i}{\hbar} \Big(\phi^{(1a)}_{jp_{j'}}(t,\beta) - \phi^{\dagger(1a)}_{jp_{j'}}(t,\beta) \Big) \Big]
\end{aligned}
$$

$$
\begin{aligned}
&= -\frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel\cdot\mathbf{R}_j^\parallel} \tilde{V}(|\mathbf{Q}^\parallel|, z_0 - R^z_{p_j}) \frac{\partial \tilde{V}(|\mathbf{Q}^\parallel + \mathbf{G}^y|, z')}{\partial z'}\bigg|_{z'=z_0-R^z_{p_{j'}}} \times \\
&\Bigg[\frac{2\,i\,W_1(\mathbf{Q}^\parallel + \mathbf{G}^y, p_{j'})}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \Bigg(e^{i\mathbf{k}\cdot\mathbf{R}_j^\parallel} \Phi_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{k}, \lambda, t, \beta) \\
&\qquad\qquad - e^{-i\mathbf{k}\cdot\mathbf{R}_j^\parallel} \Phi^\dagger_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{k}, \lambda, t, \beta) \Bigg) \\
&- \frac{i}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \Bigg(e^{i\mathbf{k}\cdot\mathbf{R}_j^\parallel} \Phi^{(1a)}_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{k}, \lambda, t, \beta) - e^{-i\mathbf{k}\cdot\mathbf{R}_j^\parallel} \Phi^{\dagger(1a)}_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{k}, \lambda, t, \beta) \Bigg) \Bigg],
\end{aligned}
\tag{E.9}
$$

where we neglected $\phi\phi^{(1a)}$ and $\phi^\dagger\phi^{\dagger(1a)}$ terms.

$\Phi^{(1a)}_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{k}, \lambda, t, \beta)$ are dimensionally a Length/(Energy·Time) and they are defined as:

$$
\begin{aligned}
\Phi^{(1a)}_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{k}, \lambda, t, \beta) \equiv \frac{1}{2m\omega_\lambda(\mathbf{k})} \mathbf{Q}^\parallel \cdot \boldsymbol{\epsilon}_{\lambda,p_j}(\mathbf{k}) \epsilon^z_{\lambda,p_{j'}}(-\mathbf{k}) \times \\
[\cos(\omega_\lambda(\mathbf{k})t)(1 + 2n_\lambda(\mathbf{k})) - i\sin(\omega_\lambda(\mathbf{k})t)]\,.
\end{aligned}
\tag{E.10}
$$

Applying the thermodynamic limit and the *Dirac comb*, we obtain:

$$
\begin{aligned}
&\frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel\cdot\mathbf{R}_j^\parallel} L^{(A1)}_{j p_{j'}} \\
&= -\frac{1}{a^2} \tilde{V}(|\mathbf{Q}^\parallel|, z_0 - R^z_{p_j}) \frac{\partial \tilde{V}(|\mathbf{Q}^\parallel + \mathbf{G}^y|, z')}{\partial z'}\bigg|_{z'=z_0-R^z_{p_{j'}}} \times \\
&\Bigg[2\,i\,W_1(\mathbf{Q}^\parallel + \mathbf{G}^y, p_{j'}) \sum_{\lambda} \Bigg(\Phi_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{Q}^\parallel, \lambda, t, \beta) \\
&\qquad\qquad - \Phi^\dagger_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, -\mathbf{Q}^\parallel, \lambda, t, \beta) \Bigg) \\
&- i \sum_{\lambda} \Bigg(\Phi^{(1a)}_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel, \lambda, t, \beta) - \Phi^{\dagger(1a)}_{p_j p_{j'}}(\mathbf{Q}^\parallel, -\mathbf{Q}^\parallel, \lambda, t, \beta) \Bigg) \Bigg] \\
&\equiv \mathscr{L}^{(A1)}_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, t, z_0, \beta)\,.
\end{aligned}
\tag{E.11}
$$

Likewise, we derive:

$$\frac{1}{a^2}\lim_{N_{xy}\to\infty}\sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel\cdot\mathbf{R}_j^\parallel} L_{jp_{j'}}^{(B1)}$$
$$= -\frac{1}{a^2}\frac{\partial\tilde{V}(|\mathbf{Q}^\parallel|,z)}{\partial z}\bigg|_{z=z_0-R^z_{p_j}} \tilde{V}(|\mathbf{Q}^\parallel+\mathbf{G}^y|,z_0-R^z_{p_{j'}})\times$$
$$\bigg[-2\,i\,W_1(\mathbf{Q}^\parallel,p_j)\sum_\lambda\bigg(\Phi_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,\mathbf{Q}^\parallel,\lambda,t,\beta)$$
$$-\Phi^\dagger_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,-\mathbf{Q}^\parallel,\lambda,t,\beta)\bigg)$$
$$+\,i\sum_\lambda\bigg(\Phi^{(1b)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,\mathbf{Q}^\parallel,\lambda,t,\beta)$$
$$-\Phi^{\dagger(1b)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,-\mathbf{Q}^\parallel,\lambda,t,\beta)\bigg)\bigg]$$
$$\equiv \mathscr{L}^{(B1)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,t,z_0,\beta)\,, \tag{E.12}$$

where we define:

$$\Phi^{(1b)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,\mathbf{k},\lambda,t,\beta)\equiv\frac{1}{2m\omega_\lambda(\mathbf{k})}\epsilon^z_{\lambda,p_j}(\mathbf{k})(\mathbf{Q}^\parallel+\mathbf{G}^y)\cdot\boldsymbol{\epsilon}_{\lambda,p_{j'}}(-\mathbf{k})\times$$
$$[\cos(\omega_\lambda(\mathbf{k})t)(1+2n_\lambda(\mathbf{k}))-i\sin(\omega_\lambda(\mathbf{k})t)]\,. \tag{E.13}$$

$\Phi^{(1b)}_{p_jp_{j'}}$ are dimensionally a Length/(Energy·Time).

Finally, we calculate the third term of Eq. (E.1) applying the one-phonon approximation. We start with:

$$\frac{1}{a^2}\lim_{N_{xy}\to\infty}\sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel\cdot\mathbf{R}_j^\parallel} L_{jp_{j'}}^{(A2)}$$
$$=\frac{1}{a^2}\lim_{N_{xy}\to\infty}\sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel\cdot\mathbf{R}_j^\parallel}\frac{1}{2}\tilde{V}(|\mathbf{Q}^\parallel|,z_0-R^z_{p_j})\times$$
$$\frac{\partial^2\tilde{V}(|\mathbf{Q}^\parallel+\mathbf{G}^y|,z')}{\partial z'^2}\bigg|_{z'=z_0-R^z_{p_{j'}}}\times$$
$$\frac{1}{\hbar}\bigg\{e^{\phi_{jj'}(t,\beta)}\bigg[-\bigg(2W_1(\mathbf{Q}^\parallel+\mathbf{G}^y,p_{j'})-\phi^{(1a)}_{jj'}(t,\beta)\bigg)^2+2W_2(p_{j'})\bigg]$$
$$-\,e^{\phi^\dagger_{jj'}(t,\beta)}\bigg[-\bigg(2W_1(\mathbf{Q}^\parallel+\mathbf{G}^y,p_{j'})-\phi^{\dagger(1a)}_{jj'}(t,\beta)\bigg)^2+2W_2(p_{j'})\bigg]\bigg\}$$

$$
\begin{aligned}
&= \frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel \cdot \mathbf{R}_j^\parallel} \frac{1}{2} \tilde{V}(|\mathbf{Q}^\parallel|, z_0 - R_{p_j}^z) \times \\
&\left. \frac{\partial^2 \tilde{V}(|\mathbf{Q}^\parallel + \mathbf{G}^y|, z')}{\partial z'^2} \right|_{z'=z_0 - R^z_{p_{j'}}} \times \\
&\Bigg[\frac{-4W_1^2(\mathbf{Q}^\parallel + \mathbf{G}^y, p_{j'}) + 2W_2(p_{j'})}{\hbar} \Big(\phi_{jp_{j'}}(t,\beta) - \phi^\dagger_{jp_{j'}}(t,\beta) \Big) \\
&+ \frac{4W_1(\mathbf{Q}^\parallel + \mathbf{G}^y, p_{j'})}{\hbar} \Big(\phi^{(1a)}_{jp_{j'}}(t,\beta) - \phi^{\dagger(1a)}_{jp_{j'}}(t,\beta) \Big) \Bigg] \\
&= \frac{1}{a^2} \lim_{N_{xy}\to\infty} \sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel \cdot \mathbf{R}_j^\parallel} \frac{1}{2} \tilde{V}(|\mathbf{Q}^\parallel|, z_0 - R_{p_j}^z) \times \\
&\left. \frac{\partial^2 \tilde{V}(|\mathbf{Q}^\parallel + \mathbf{G}^y|, z')}{\partial z'^2} \right|_{z'=z_0 - R^z_{p_{j'}}} \Bigg[\frac{-4W_1^2(\mathbf{Q}^\parallel + \mathbf{G}^y, p_{j'}) + 2W_2(p_{j'})}{N_{xy}} \times \\
&\sum_{\mathbf{k},\lambda}^{BZ} \Big(e^{i\mathbf{k}\cdot\mathbf{R}_j^\parallel} \Phi_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{k}, \lambda, t, \beta) \\
&\qquad\qquad - e^{-i\mathbf{k}\cdot\mathbf{R}_j^\parallel} \Phi^\dagger_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{k}, \lambda, t, \beta) \Big) \\
&+ \frac{4W_1(\mathbf{Q}^\parallel + \mathbf{G}^y, p_{j'})}{N_{xy}} \sum_{\mathbf{k},\lambda}^{BZ} \Big(e^{i\mathbf{k}\cdot\mathbf{R}_j^\parallel} \Phi^{(1a)}_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{k}, \lambda, t, \beta) \\
&\qquad\qquad - e^{-i\mathbf{k}\cdot\mathbf{R}_j^\parallel} \Phi^{\dagger(1a)}_{p_j p_{j'}}(\mathbf{Q}^\parallel, \mathbf{k}, \lambda, t, \beta) \Big) \Bigg],
\end{aligned}
\tag{E.14}
$$

where we neglected $\phi\phi^{(1a)}$, $\phi^\dagger\phi^{\dagger(1a)}$, $\left(\phi^{(1a)}\right)^2$ and $\left(\phi^{\dagger(1a)}\right)^2$ terms.

Applying the thermodynamic limit and the *Dirac comb*, we obtain:

$$\frac{1}{a^2}\lim_{N_{xy}\to\infty}\sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel\cdot\mathbf{R}_j^\parallel} L^{(A2)}_{jp_{j'}}$$
$$= \frac{1}{2a^2}\tilde{V}(|\mathbf{Q}^\parallel|, z_0 - R^z_{p_j})\frac{\partial^2\tilde{V}(|\mathbf{Q}^\parallel + \mathbf{G}^y|, z')}{\partial z'^2}\bigg|_{z'=z_0-R^z_{p_{j'}}}\times$$
$$\Bigg[\bigg(-4W_1^2(\mathbf{Q}^\parallel + \mathbf{G}^y, p_{j'}) + 2W_2(p_{j'})\bigg)\sum_\lambda\bigg(\Phi_{p_jp_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{Q}^\parallel, \lambda, t, \beta)$$
$$- \Phi^\dagger_{p_jp_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, -\mathbf{Q}^\parallel, \lambda, t, \beta)\bigg) + 4W_1(\mathbf{Q}^\parallel + \mathbf{G}^y, p_{j'})\times$$
$$\sum_\lambda\bigg(\Phi^{(1a)}_{p_jp_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel, \lambda, t, \beta) - \Phi^{\dagger(1a)}_{p_jp_{j'}}(\mathbf{Q}^\parallel, -\mathbf{Q}^\parallel, \lambda, t, \beta)\bigg)\Bigg]$$
$$\equiv \mathscr{L}^{(A2)}_{p_jp_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, t, z_0, \beta)\,. \tag{E.15}$$

Similarly, we derive:

$$\frac{1}{a^2}\lim_{N_{xy}\to\infty}\sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel\cdot\mathbf{R}_j^\parallel} L^{(B2)}_{jp_{j'}}$$
$$= \frac{1}{2a^2}\frac{\partial^2\tilde{V}(|\mathbf{Q}^\parallel|, z)}{\partial z^2}\bigg|_{z=z_0-R^z_{p_j}}\tilde{V}(|\mathbf{Q}^\parallel + \mathbf{G}^y|, z_0 - R^z_{p_{j'}})\times$$
$$\Bigg[\bigg(-4W_1^2(\mathbf{Q}^\parallel, p_j) + 2W_2(p_j)\bigg)\sum_\lambda\bigg(\Phi_{p_jp_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{Q}^\parallel, \lambda, t, \beta)$$
$$- \Phi^\dagger_{p_jp_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, -\mathbf{Q}^\parallel, \lambda, t, \beta)\bigg) + 4W_1(\mathbf{Q}^\parallel, p_j)\times$$
$$\sum_\lambda\bigg(\Phi^{(1b)}_{p_jp_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, \mathbf{Q}^\parallel, \lambda, t, \beta)$$
$$- \Phi^{\dagger(1b)}_{p_jp_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, -\mathbf{Q}^\parallel, \lambda, t, \beta)\bigg)\Bigg]$$
$$\equiv \mathscr{L}^{(B2)}_{p_jp_{j'}}(\mathbf{Q}^\parallel, \mathbf{Q}^\parallel + \mathbf{G}^y, t, z_0, \beta)\,, \tag{E.16}$$

and

$$\frac{1}{a^2}\lim_{N_{xy}\to\infty}\sum_{j_\parallel} e^{-i\mathbf{Q}^\parallel\cdot\mathbf{R}_j^\parallel} L_{jp_{j'}}^{(C2)}$$
$$= \frac{1}{a^2}\frac{\partial\tilde{V}(|\mathbf{Q}^\parallel|,z)}{\partial z}\bigg|_{z=z_0-R^z_{p_j}} \frac{\partial\tilde{V}(|\mathbf{Q}^\parallel+\mathbf{G}^y|,z')}{\partial z'}\bigg|_{z'=z_0-R^z_{p_{j'}}}\times$$
$$\Bigg[4W_1(\mathbf{Q}^\parallel,p_j)W_1(\mathbf{Q}^\parallel+\mathbf{G}^y,p_{j'})\sum_\lambda\Big(\Phi_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,\mathbf{Q}^\parallel,\lambda,t,\beta)$$
$$-\Phi^\dagger_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,-\mathbf{Q}^\parallel,\lambda,t,\beta)\Big)$$
$$-2W_1(\mathbf{Q}^\parallel,p_j)\sum_\lambda\Big(\Phi^{(1a)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel,\lambda,t,\beta)-\Phi^{\dagger(1a)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,-\mathbf{Q}^\parallel,\lambda,t,\beta)\Big)$$
$$-2W_1(\mathbf{Q}^\parallel+\mathbf{G}^y,p_{j'})\sum_\lambda\Big(\Phi^{(1b)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,\mathbf{Q}^\parallel,\lambda,t,\beta)$$
$$-\Phi^{\dagger(1b)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,-\mathbf{Q}^\parallel,\lambda,t,\beta)\Big)$$
$$+\sum_\lambda\Big(\Phi^{(2)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel,\lambda,t,\beta)-\Phi^{\dagger(2)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,-\mathbf{Q}^\parallel,\lambda,t,\beta)\Big)\Bigg]$$
$$\equiv \mathscr{L}^{(C2)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,t,z_0,\beta)\,, \tag{E.17}$$

where we define:

$$\Phi^{(2)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{k},\lambda,t,\beta)\equiv\frac{1}{2m\omega_\lambda(\mathbf{k})}\epsilon^z_{\lambda,p_j}(\mathbf{k})\epsilon^z_{\lambda,p_{j'}}(-\mathbf{k})\times$$
$$[\cos(\omega_\lambda(\mathbf{k})t)(1+2n_\lambda(\mathbf{k}))-i\sin(\omega_\lambda(\mathbf{k})t)]\,. \tag{E.18}$$

$\Phi^{(2)}_{p_jp_{j'}}$ are dimensionally an Area/(Energy·Time).

Then, we have all the elements to simplify Eq. (E.1):

$$F=\sum_{\mathbf{G}^y}e^{-iG^yy_0}\int\frac{d^2Q^\parallel}{(2\pi)^2}\mathbf{Q}^\parallel\cdot\hat{\mathbf{v}}_{\mathrm{SL}}\ \mathrm{Re}\Bigg(\sum_{p_j,p_{j'}}e^{-W(\mathbf{Q}^\parallel,p_j)}e^{-W(\mathbf{Q}^\parallel+\mathbf{G}^y,p_{j'})}\times$$
$$\int_0^\infty dt\,e^{(i\mathbf{Q}^\parallel\cdot\mathbf{v}_{\mathrm{SL}}-\gamma/2)t}\Big(\mathscr{L}^{(0)}_{p_jp_{j'}}+\mathscr{L}^{(1)}_{p_jp_{j'}}+\mathscr{L}^{(2)}_{p_jp_{j'}}\Big)\Bigg)\,, \tag{E.19}$$

where $\mathscr{L}^{(0)}_{p_jp_{j'}}$ is defined in Eq. (E.8), and

$$\mathscr{L}^{(1)}_{p_jp_{j'}}=\mathscr{L}^{(A1)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,t,z_0,\beta)+\mathscr{L}^{(B1)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,t,z_0,\beta)\,, \tag{E.20}$$

$$\mathscr{L}^{(2)}_{p_jp_{j'}}=\mathscr{L}^{(A2)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,t,z_0,\beta)+\mathscr{L}^{(B2)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,t,z_0,\beta)$$
$$+\mathscr{L}^{(C2)}_{p_jp_{j'}}(\mathbf{Q}^\parallel,\mathbf{Q}^\parallel+\mathbf{G}^y,t,z_0,\beta)\,. \tag{E.21}$$

Here, the ingredients $\mathscr{L}^{(A1)}_{p_j p_{j'}}$ and $\mathscr{L}^{(B1)}_{p_j p_{j'}}$ are defined in Eq. (E.11) and in Eq. (E.12), respectively. $\mathscr{L}^{(A2)}_{p_j p_{j'}}$, $\mathscr{L}^{(B2)}_{p_j p_{j'}}$ and $\mathscr{L}^{(C2)}_{p_j p_{j'}}$ are defined in Eq. (E.15), in Eq. (E.16), and in Eq. (E.17), respectively.

$\mathscr{L}^{(k)}_{p_j p_{j'}}$ are functions of $(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\parallel} + \mathbf{G}^{y}, t, z_0, \beta)$ describing the effects of lattice vibration to k^{th} order in the out-of-plane displacements $\hat{u}^{z}$, in the one-phonon approximation.
The $\mathscr{L}^{(k)}_{p_j p_{j'}}$ functions are dimensionally an Energy·Area/Time.

Appendix F

The dynamical matrix

In this Appendix we evaluate the dynamical matrix for the evaluation of the harmonic vibrations of the 2D square-lattice network and for the 3D simple-cubic crystal.

F.1 2D

We start with the calculation of the dynamical matrix elements of a 2D harmonic crystal by particles of mass m that at equilibrium are arranged as a square lattice. Harmonic first- and second-neighbor springs with elastic constant K and K' and equilibrium lengths a and $\sqrt{2}a$, respectively, guarantee the mechanical stability (see Figure F.1).

The potential energy V_{tot} of our problem can be expressed as

$$V_{\text{tot}} = \frac{1}{2}\sum_{j=1}^{N} V_{\text{tot}}^{j} \, , \tag{F.1}$$

with

$$V_{\text{tot}}^{j} = \sum_{\langle j,j'\rangle_1} \frac{K}{2}(r_{jj'} - a)^2 + \sum_{\langle j,j'\rangle_2} \frac{K'}{2}(r_{jj'} - \sqrt{2}a)^2 \equiv V_{\text{tot}}^{j(\text{1st})} + V_{\text{tot}}^{j(\text{2nd})} \, , \tag{F.2}$$

where $r_{jj'} \equiv \sqrt{(x_j - x_{j'})^2 + (y_j - y_{j'})^2}$ is the distance between two atoms, $\langle \cdot, \cdot \rangle_1$ and $\langle \cdot, \cdot \rangle_2$ indicate the first- and second-neighbor pairs respectively and N is the number of atoms of our system.

The dynamical matrix elements $D_{\mu\nu}(\mathbf{q})$ of the crystal in reciprocal space are defined as

$$D_{\mu\nu}(\mathbf{q}) = \sum_{j'} D_{j\mu,j'\nu} e^{-i\mathbf{q}\cdot(\mathbf{R}_j - \mathbf{R}_{j'})} \, , \tag{F.3}$$

where the sum runs over all the atoms of the crystal, $\mathbf{R}_j$ and $\mathbf{R}_{j'}$ are the equilibrium positions of the j-th and j'-th particles respectively and $\mathbf{q}$ spans

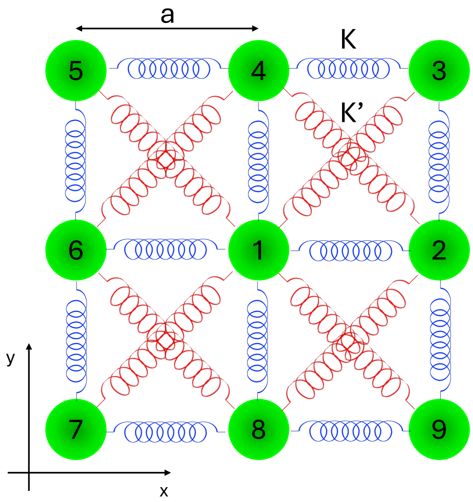

FIGURE F.1: Graphical representation of a 3×3 portion of the 2D harmonic crystal. Each atom interacts with first-neighbor atoms through springs with elastic constant K (coloured in blue) and with second-neighbor atoms through springs with elastic constant K' (coloured in red).

the first Brillouin zone. $D_{j\mu,j'\nu}$ is the "interatomic force constants" defined as the second derivative of the potential V_{tot} evaluated at the equilibrium configuration:

$$D_{j\mu,j'\nu} = \frac{1}{m}\left(\frac{\partial^2 V_{\text{tot}}}{\partial u_{j\mu}\,\partial u_{j'\nu}}\right)_0, \tag{F.4}$$

where $u_{j\mu}$ is the displacement of the j-th particle from equilibrium position along μ direction [22].

We start to consider the interaction with first-neighbor atoms around the $j = 1$ particle. We label the atoms as shown in Figure F.1. The potential energy $V_{\text{tot}}^{j=1(1\text{st})}$ is

$$\begin{aligned} V_{\text{tot}}^{j=1(1\text{st})} =& \frac{K}{2}\Bigg\{\left[\sqrt{\left(a + u_{2x} - u_{1x}\right)^2 + \left(u_{2y} - u_{1y}\right)^2} - a\right]^2 \\ &+ \left[\sqrt{\left(a + u_{1x} - u_{6x}\right)^2 + \left(u_{1y} - u_{6y}\right)^2} - a\right]^2 \\ &+ \left[\sqrt{\left(u_{4x} - u_{1x}\right)^2 + \left(a + u_{4y} - u_{1y}\right)^2} - a\right]^2 \\ &+ \left[\sqrt{\left(u_{1x} - u_{8x}\right)^2 + \left(a + u_{1y} - u_{8y}\right)^2} - a\right]^2\Bigg\}. \end{aligned} \tag{F.5}$$

The first derivative $\frac{\partial V_{\rm tot}^{j=1(1{\rm st})}}{\partial u_{1x}}$ is

$$\begin{aligned}\frac{\partial V_{\rm tot}^{j=1(1{\rm st})}}{\partial u_{1x}} =&K\Bigg\{-\frac{\Big[\sqrt{\big(a+u_{2x}-u_{1x}\big)^2+\big(u_{2y}-u_{1y}\big)^2}-a\Big]\big(a+u_{2x}-u_{1x}\big)}{r_{12}}\\ &+\frac{\Big[\sqrt{\big(a+u_{1x}-u_{6x}\big)^2+\big(u_{1y}-u_{6y}\big)^2}-a\Big]\big(a+u_{1x}-u_{6x}\big)}{r_{16}}\\ &-\frac{\Big[\sqrt{\big(u_{4x}-u_{1x}\big)^2+\big(a+u_{4y}-u_{1y}\big)^2}-a\Big]\big(u_{4x}-u_{1x}\big)}{r_{14}}\\ &+\frac{\Big[\sqrt{\big(u_{1x}-u_{8x}\big)^2+\big(a+u_{1y}-u_{8y}\big)^2}-a\Big]\big(u_{1x}-u_{8x}\big)}{r_{18}}\Bigg\}.\end{aligned} \tag{F.6}$$

Now we can calculate $D_{1x,j'\nu}$ where $j'=2,4,6,8$ and $\nu=x,y$. The only non-zero terms are

$$\begin{aligned}D_{1x,2x} =&\frac{1}{m}\left(\frac{\partial^2 V_{\rm tot}^{j=1(1{\rm st})}}{\partial u_{1x}\,\partial u_{2x}}\right)_0\\ =&-\frac{K}{m}\left[\frac{\big(a+u_{2x}-u_{1x}\big)^2}{\big(a+u_{2x}-u_{1x}\big)^2+\big(u_{2y}-u_{1y}\big)^2}\right]_0=-\frac{K}{m}\,,\end{aligned} \tag{F.7}$$

$$\begin{aligned}D_{1x,6x} =&\frac{1}{m}\left(\frac{\partial^2 V_{\rm tot}^{j=1(1{\rm st})}}{\partial u_{1x}\,\partial u_{6x}}\right)_0\\ =&-\frac{K}{m}\left[\frac{\big(a+u_{1x}-u_{6x}\big)^2}{\big(a+u_{1x}-u_{6x}\big)^2+\big(u_{1y}-u_{6y}\big)^2}\right]_0=-\frac{K}{m}\,.\end{aligned} \tag{F.8}$$

Similarly we can calculate $D_{1y,j'\nu}$ where $j'=2,4,6,8$ and $\nu=x,y$. The only non-zero terms are

$$D_{1y,4y}=D_{1y,8y}=-\frac{K}{m}\,. \tag{F.9}$$

All $D_{1x,j'y}$ and $D_{1y,j'x}$ terms vanish.

Now we consider the interaction with second-neighbor atoms around the

$j = 1$ particle. The potential energy $V_{\text{tot}}^{j=1(\text{2nd})}$ is

$$\begin{aligned} V_{\text{tot}}^{j=1(\text{2nd})} =& \frac{K'}{2}\Bigg\{\left[\sqrt{\left(a+u_{3x}-u_{1x}\right)^2+\left(a+u_{3y}-u_{1y}\right)^2}-a\sqrt{2}\right]^2 \\ &+\left[\sqrt{\left(a+u_{1x}-u_{5x}\right)^2+\left(a+u_{5y}-u_{1y}\right)^2}-a\sqrt{2}\right]^2 \\ &+\left[\sqrt{\left(a+u_{1x}-u_{7x}\right)^2+\left(a+u_{1y}-u_{7y}\right)^2}-a\sqrt{2}\right]^2 \\ &+\left[\sqrt{\left(a+u_{9x}-u_{1x}\right)^2+\left(a+u_{1y}-u_{9y}\right)^2}-a\sqrt{2}\right]^2\Bigg\}. \end{aligned} \tag{F.10}$$

The first derivative $\frac{\partial V_{\text{tot}}^{j=1(\text{2nd})}}{\partial u_{1x}}$ is

$$\begin{aligned} &\frac{\partial V_{\text{tot}}^{j=1(\text{2nd})}}{\partial u_{1x}} = \\ &K'\Bigg\{-\frac{\left[\sqrt{\left(a+u_{3x}-u_{1x}\right)^2+\left(a+u_{3y}-u_{1y}\right)^2}-a\sqrt{2}\right]\left(a+u_{3x}-u_{1x}\right)}{r_{13}} \\ &+\frac{\left[\sqrt{\left(a+u_{1x}-u_{5x}\right)^2+\left(a+u_{5y}-u_{1y}\right)^2}-a\sqrt{2}\right]\left(a+u_{1x}-u_{5x}\right)}{r_{15}} \\ &+\frac{\left[\sqrt{\left(a+u_{1x}-u_{7x}\right)^2+\left(a+u_{1y}-u_{7y}\right)^2}-a\sqrt{2}\right]\left(a+u_{1x}-u_{7x}\right)}{r_{17}} \\ &-\frac{\left[\sqrt{\left(a+u_{9x}-u_{1x}\right)^2+\left(a+u_{1y}-u_{9y}\right)^2}-a\sqrt{2}\right]\left(a+u_{9x}-u_{1x}\right)}{r_{19}}\Bigg\}. \end{aligned} \tag{F.11}$$

We start to calculate $D_{1x,3x}$:

$$\begin{aligned} D_{1x,3x} =& \frac{1}{m}\left(\frac{\partial^2 V_{\text{tot}}^{j=1(\text{2nd})}}{\partial u_{1x}\,\partial u_{3x}}\right)_0 \\ =& -\frac{K'}{m}\left[\frac{\left(a+u_{3x}-u_{1x}\right)^2}{\left(a+u_{3x}-u_{1x}\right)^2+\left(a+u_{3y}-u_{1y}\right)^2}\right]_0 = -\frac{K'}{2m}\,. \end{aligned} \tag{F.12}$$

Likewise we obtain

$$D_{1x,5x} = D_{1x,7x} = D_{1x,9x} = -\frac{K'}{2m}\,, \tag{F.13}$$

and

$$D_{1y,3y} = D_{1y,5y} = D_{1y,7y} = D_{1y,9y} = -\frac{K'}{2m}\,. \tag{F.14}$$

Now we proceed to calculate $D_{1x,3y}$:

$$\begin{aligned}D_{1x,3y} =&\frac{1}{m}\left(\frac{\partial^2 V_{\text{tot}}^{j=1(\text{2nd})}}{\partial u_{1x}\,\partial u_{3y}}\right)_0\\ =&-\frac{K'}{m}\left[\frac{\big(a+u_{3x}-u_{1x}\big)\big(a+u_{3y}-u_{1y}\big)}{\big(a+u_{3x}-u_{1x}\big)^2+\big(a+u_{3y}-u_{1y}\big)^2}\right]_0 = -\frac{K'}{2m}\\ =&D_{1y,3x}\,. \end{aligned} \tag{F.15}$$

Likewise we have

$$D_{1x,5y} = D_{1y,5x} = \frac{K'}{2m}\,, \tag{F.16}$$

$$D_{1x,7y} = D_{1y,7x} = -\frac{K'}{2m}\,, \tag{F.17}$$

$$D_{1x,9y} = D_{1y,9x} = \frac{K'}{2m}\,. \tag{F.18}$$

Finally we evaluate $D_{1x,1x}$:

$$\begin{aligned}D_{1x,1x} =&\frac{1}{m}\left(\frac{\partial^2 V_{\text{tot}}^{j=1(\text{1st})}}{\partial u_{1x}\,\partial u_{1x}}\right)_0 + \frac{1}{m}\left(\frac{\partial^2 V_{\text{tot}}^{j=1(\text{2nd})}}{\partial u_{1x}\,\partial u_{1x}}\right)_0\\ =&\frac{K}{m}\left[\frac{\big(a+u_{2x}-u_{1x}\big)^2}{\big(a+u_{2x}-u_{1x}\big)^2+\big(u_{2y}-u_{1y}\big)^2}\right]_0\\ &+\frac{K}{m}\left[\frac{\big(a+u_{1x}-u_{6x}\big)^2}{\big(a+u_{1x}-u_{6x}\big)^2+\big(u_{1y}-u_{6y}\big)^2}\right]_0\\ &+\frac{4K'}{m}\left[\frac{\big(a+u_{3x}-u_{1x}\big)^2}{\big(a+u_{3x}-u_{1x}\big)^2+\big(a+u_{3y}-u_{1y}\big)^2}\right]_0 = \frac{2K}{m}+\frac{2K'}{m}\,. \end{aligned} \tag{F.19}$$

Through the same derivation we obtain:

$$D_{1y,1y} = \frac{2K}{m} + \frac{2K'}{m}\,. \tag{F.20}$$

Now we all the ingredients to calculate the dynamical matrix elements using Eq. (F.3). We start from the diagonal terms:

$$\begin{aligned}
D_{xx}(\mathbf{q}) =& \frac{2K}{m} - \frac{K}{m} e^{-iq_x(-a)} - \frac{K}{m} e^{-iq_x(a)} + \frac{2K'}{m} \\
& - \frac{K'}{2m} \Big[e^{-iq_x(-a)-iq_y(-a)} + e^{-iq_x(a)-iq_y(-a)} + e^{-iq_x(a)-iq_y(a)} \\
& + e^{-iq_x(-a)-iq_y(a)} \Big] \\
=& \frac{2K}{m} \Big[1 - \cos(q_x a) \Big] + \frac{2K'}{m} - \frac{K'}{2m} \Big[e^{iq_x a} \Big(e^{-iq_y a} + e^{iq_y a} \Big) \\
& + e^{-iq_x a} \Big(e^{-iq_y a} + e^{iq_y a} \Big) \Big] \\
=& \frac{2K}{m} \Big[1 - \cos(q_x a) \Big] + \frac{2K'}{m} - \frac{K'}{2m} \Big(e^{iq_x a} + e^{-iq_x a} \Big) \Big(e^{-iq_y a} + e^{iq_y a} \Big) \\
=& \frac{2K}{m} \Big[1 - \cos(q_x a) \Big] + \frac{2K'}{m} \Big[1 - \cos(q_x a) \cos(q_y a) \Big] , \qquad \text{(F.21)}
\end{aligned}$$

and likewise we obtain

$$D_{yy}(\mathbf{q}) = \frac{2K}{m} \Big[1 - \cos(q_y a) \Big] + \frac{2K'}{m} \Big[1 - \cos(q_y a) \cos(q_x a) \Big] . \qquad \text{(F.22)}$$

Finally we calculate the off-diagonal terms:

$$\begin{aligned}
D_{xy}(\mathbf{q}) =& - \frac{K'}{2m} \Big[e^{-iq_x(-a)-iq_y(-a)} - e^{-iq_x(a)-iq_y(-a)} \\
& + e^{-iq_x(a)-iq_y(a)} - e^{-iq_x(-a)-iq_y(a)} \Big] \\
=& - \frac{K'}{2m} \Big[e^{iq_x a} \Big(e^{iq_y a} - e^{-iq_y a} \Big) - e^{-iq_x a} \Big(e^{iq_y a} - e^{-iq_y a} \Big) \Big] \\
=& - \frac{K'}{2m} \Big(e^{iq_x a} - e^{-iq_x a} \Big) \Big(e^{iq_y a} - e^{-iq_y a} \Big) \\
=& \frac{2K'}{m} \sin(q_x a) \sin(q_y a) = D_{yx}(\mathbf{q}) . \qquad \text{(F.23)}
\end{aligned}$$

The phonon polarization vectors $\boldsymbol{\epsilon}_\lambda(\mathbf{q})$ and frequencies $\omega_\lambda(\mathbf{q})$ are obtained by diagonalizing the dynamical matrix $D_{\mu\nu}(\mathbf{q})$. From the secular equation

$$\omega_\lambda^2(\mathbf{q}) \epsilon_{\lambda\mu}(\mathbf{q}) = \sum_\nu D_{\mu\nu}(\mathbf{q}) \epsilon_{\lambda\nu}(\mathbf{q}) , \qquad \text{(F.24)}$$

we obtain the dispersion relations:

$$\omega_{\pm}(\mathbf{q}) = \Bigg\{ 2\frac{K}{m}\left[\sin^2\left(\frac{q_x a}{2}\right) + \sin^2\left(\frac{q_y a}{2}\right) + \frac{K'}{K}\Big(1 - \cos(q_x a)\cos(q_y a)\Big)\right]$$
$$\pm\, 2\frac{K}{m}\left[\left(\sin^2\left(\frac{q_x a}{2}\right) - \sin^2\left(\frac{q_y a}{2}\right)\right)^2 + \left(\frac{K'}{K}\sin(q_x a)\sin(q_y a)\right)^2\right]^{1/2}\Bigg\}^{1/2}, \tag{F.25}$$

for the $\lambda = 1$ transverse $(-)$ and $\lambda = 2$ longitudinal $(+)$ phonons.

F.2 3D

Now, let us consider a 3D harmonic crystal characterized by particles of mass m that at equilibrium are arranged as a simple-cubic lattice. Even in this case, harmonic nearest- and second-neighbor springs with elastic constant K and K' and equilibrium lengths a and $\sqrt{2}a$, respectively, guarantee the mechanical stability. Relations (F.1), (F.2), (F.3) and (F.4) continue to be valid and the distance between two atoms $r_{jj'}$ is expressed as

$$r_{jj'} = \sqrt{(x_j - x_{j'})^2 + (y_j - y_{j'})^2 + (z_j - z_{j'})^2}\,. \tag{F.26}$$

Iterating the 2D model technique, the 3×3 3D dynamical matrix can be expressed as an explicit function of $\mathbf{q}$. The diagonal elements are:

$$D_{\mu\mu}(\mathbf{q}) = \frac{2K}{m}\Big[1 - \cos(q_\mu a)\Big] + \frac{2K'}{m}\Big[2 - \cos(q_\mu a)(\cos(q_\nu a) + \cos(q_\gamma a))\Big], \tag{F.27}$$

with $\mu,\, \nu,\, \gamma = x,\, y,\, z, \quad \mu \neq \nu,\, \nu \neq \gamma$ and $\mu \neq \gamma$.

The off-diagonal elements are:

$$D_{\mu\nu}(\mathbf{q}) = \frac{2K'}{m}\sin(q_\mu a)\sin(q_\nu a)\,, \tag{F.28}$$

with $\mu,\, \nu = x,\, y,\, z$ and $\mu \neq \nu$.

F.3 3D crystal slab

Now we proceed to analyze our case of interest: a 3D harmonic semi-infinite crystal where at equilibrium the particles of mass m are arranged as a simple-cubic lattice truncated at a (001) surface. Harmonic nearest- and second-neighbor springs with, respectively, elastic constant K and K' have equilibrium lengths a and $\sqrt{2}a$. This system retains a discrete translational symmetry along xy-plane; therefore, the wave-vector $\mathbf{q}$ spans the first Brillouin zone of the 2D square lattice in the x and y directions.

For our purpose let us consider a slab composed of N_p layers where $N_p \to \infty$. There are N_p atoms in each primitive cell and the p-th atom of the cell is in the position $(0, 0, -a(p-1))$ with $p = 1, 2, ..., N_p$. We need to generalize the dynamical matrix elements of Eq. (F.3) for a crystal with a basis:

$$D_{p\mu,p'\nu}(\mathbf{q}) = \sum_{n'} D_{np\mu,n'p'\nu} e^{-i\mathbf{q}\cdot(\mathbf{R}_n - \mathbf{R}_{n'})}\,, \tag{F.29}$$

where the sum runs over all the unit cells, $\mathbf{R}_n$ and $\mathbf{R}_{n'}$ are the positions of the n-th and n'-th cells respectively and $\mathbf{q}$ spans the first Brillouin zone. The interatomic force constant $D_{np\mu,n'p'\nu}$ is always defined as the second derivative of the potential V_{tot} evaluated at the equilibrium configuration:

$$D_{np\mu,n'p'\nu} = \frac{1}{m}\left(\frac{\partial^2 V_{\text{tot}}}{\partial u_{np\mu}\partial u_{n'p'\nu}}\right)_0\,, \tag{F.30}$$

where now $u_{np\mu}$ is the displacement of the p-th atom of the n-th unit cell from equilibrium position along μ direction.

Iterating the 2D model technique the $3N_p \times 3N_p$ dynamical matrix can be expressed as an explicit functions of $\mathbf{q}$. The elements are described by the following relations valid for $p = 1, 2, ..., N_p$ and for $\mu, \nu = x, y$:

$$D_{p\mu,p\mu} = \frac{2K}{m}\Big[1 - \cos(q_\mu a)\Big] + \frac{2K'}{m}\Big[1 - \cos(q_\mu a)\cos(q_\nu a)\Big] \quad \text{for } \mu \neq \nu\,, \tag{F.31}$$

$$D_{p\mu,p\nu} = \frac{2K'}{m}\sin(q_\mu a)\sin(q_\nu a) \quad \text{for } \mu \neq \nu\,, \tag{F.32}$$

$$D_{p\mu,p'\mu} = -\frac{K'}{m}\cos(q_\mu a) \quad \text{for } |p - p'| = 1\,, \tag{F.33}$$

$$D_{p\mu,p'z} = D_{pz,p'\mu} = i\frac{K'}{m}\sin(q_\mu a) \quad \text{for } p' = p + 1\,, \tag{F.34}$$

$$D_{p\mu,p'z} = D_{pz,p'\mu} = -i\frac{K'}{m}\sin(q_\mu a) \quad \text{for } p' = p - 1\,, \tag{F.35}$$

$$D_{pz,p'z} = -\frac{K}{m} - \frac{K'}{m}\Big[\cos(q_x a) + \cos(q_y a)\Big] \quad \text{for } |p - p'| = 1\,, \tag{F.36}$$

assuming that all terms with $p' = 0$ and $p' = N_p+1$ are absent. Furthermore, all other elements not explicitly written are null.

The $3N_p$ vibrational eigenfrequencies and the corresponding polarization vectors are then obtained by diagonalizing the resulting $3N_p \times 3N_p$ dynamical matrix.

Appendix G

Green's Function of a One-Dimensional Chain for the calculation of normal modes

We begin with a semi-infinite one dimensional chain composed of identical atoms along x-direction connected with a spring of elastic constant K. We express the position operator of the i-th atom as

$$\hat{x}_i = R_i + \hat{u}_i \,, \qquad 1 \leq i < \infty \tag{G.1}$$

where R_i is the equilibrium position and $\hat{u}_i$ is the displacement operator of the i-th atom.

Let us define a as their equilibrium distance: $a = R_{i+1} - R_i$.

The Hamiltonian of the system is

$$\hat{H} = \sum_{i=1}^{\infty} \frac{\hat{p}_i^2}{2m} + \sum_{i=1}^{\infty} \frac{1}{2} K (\hat{x}_{i+1} - \hat{x}_i - a)^2 - F(t)\,(\hat{x}_1 - R_1)\,, \tag{G.2}$$

where m and $\hat{p}_i$ are the mass and momentum operator of the i-th atom, respectively, while $\hat{x}_1$ and R_1 are the position operator and equilibrium position of the first atom. The external force $F(t)$ is applied only on the first atom. Using assumptions (G.1) the Hamiltonian can be written as

$$\hat{H} = \sum_{i=1}^{\infty} \frac{\hat{p}_i^2}{2m} + \sum_{i=1}^{\infty} \frac{1}{2} K (\hat{u}_{i+1} - \hat{u}_i)^2 - F(t)\,\hat{u}_1. \tag{G.3}$$

$\hat{u}_i$ and $\hat{p}_i$ satisfy the usual commutation rules, i.e.

$$[\hat{u}_i, \hat{p}_i] = i\hbar\delta_{i,j} \,, \tag{G.4}$$

$$[\hat{u}_i, \hat{u}_j] = [\hat{p}_i, \hat{p}_j] = 0\,. \tag{G.5}$$

In Heisenberg picture, the time evolution of the displacement and momentum operators for the first atom is given by:

$$\frac{d}{dt}\hat{u}_{1\,\mathrm{H}}(t) = \frac{i}{\hbar}[\hat{H}(t), \hat{u}_{1\,\mathrm{H}}(t)]\,, \tag{G.6}$$

$$\frac{d}{dt}\hat{p}_{1\,\mathrm{H}}(t) = \frac{i}{\hbar}[\hat{H}(t), \hat{p}_{1\,\mathrm{H}}(t)]\,. \tag{G.7}$$

We start to evaluate (G.6) using commutation rules (G.4), (G.5), and the Hamiltonian written in (G.3):

$$\begin{aligned}
\frac{d}{dt}\hat{u}_{1\,\mathrm{H}}(t) &= \frac{i}{\hbar}\hat{U}^{\dagger}(t,0)[\hat{H}(t), \hat{u}_1]\hat{U}(t,0) = \frac{i}{\hbar}\hat{U}^{\dagger}(t,0)\left[\frac{\hat{p}_1^2}{2m}, \hat{u}_1\right]\hat{U}(t,0) \\
&= \frac{i}{2m\hbar}\hat{U}^{\dagger}(t,0)\left[\hat{p}_1^2, \hat{u}_1\right]\hat{U}(t,0) = \frac{i}{2m\hbar}\hat{U}^{\dagger}(t,0)(2\hat{p}_1\left[\hat{p}_1, \hat{u}_1\right])\hat{U}(t,0) \\
&= \frac{i}{2m\hbar}\hat{U}^{\dagger}(t,0)(-2\hat{p}_1 i\hbar)\hat{U}(t,0) = \frac{1}{m}\hat{U}(t,0)\hat{p}_1\hat{U}(t,0) \\
&= \frac{1}{m}\hat{p}_{1\,\mathrm{H}}(t).
\end{aligned} \tag{G.8}$$

Now we evaluate (G.7) using the same commutation rules (G.4), (G.5) and Hamiltonian (G.3):

$$\begin{aligned}
\frac{d}{dt}\hat{p}_{1\,\mathrm{H}}(t) =& \frac{i}{\hbar}\hat{U}^{\dagger}(t,0)[\hat{H}(t), \hat{p}_1]\hat{U}(t,0) \\
=& \frac{i}{\hbar}\hat{U}^{\dagger}(t,0)\left[\frac{1}{2}K(\hat{u}_2 - \hat{u}_1)^2 - F(t)\hat{u}_1, \hat{p}_1\right]\hat{U}(t,0) \\
=& \frac{i}{\hbar}\hat{U}^{\dagger}(t,0)\left[\frac{1}{2}K\hat{u}_1^2 - K\hat{u}_1\hat{u}_2 - F(t)\hat{u}_1, \hat{p}_1\right]\hat{U}(t,0) \\
=& \frac{i}{2\hbar}K\hat{U}^{\dagger}(t,0)[\hat{u}_1^2, \hat{p}_1]\hat{U}(t,0) - \frac{i}{\hbar}K\hat{U}^{\dagger}(t,0)[\hat{u}_1\hat{u}_2, \hat{p}_1]\hat{U}(t,0) \\
&- \frac{i}{\hbar}F(t)\hat{U}^{\dagger}(t,0)[\hat{u}_1, \hat{p}_1]\hat{U}(t,0) \\
=& \frac{i}{2\hbar}K\hat{U}^{\dagger}(t,0)(2\hat{u}_1[\hat{u}_1, \hat{p}_1])\hat{U}(t,0) - \frac{i}{\hbar}K\hat{U}^{\dagger}(t,0)([\hat{u}_1, \hat{p}_1]\hat{u}_2)\hat{U}(t,0) \\
&- \frac{i}{\hbar}F(t)\hat{U}^{\dagger}(t,0)(i\hbar)\hat{U}(t,0) \\
=& \frac{i}{2\hbar}K\hat{U}^{\dagger}(t,0)(2i\hbar\hat{u}_1)\hat{U}(t,0) - \frac{i}{\hbar}K\hat{U}^{\dagger}(t,0)(i\hbar\hat{u}_2)\hat{U}(t,0) + F(t) \\
=& -K\hat{U}^{\dagger}(t,0)\hat{u}_1\hat{U}(t,0) + K\hat{U}^{\dagger}(t,0)\hat{u}_2\hat{U}(t,0) + F(t) \\
=& -K\hat{u}_{1\,\mathrm{H}}(t) + K\hat{u}_{2\,\mathrm{H}}(t) + F(t) \\
=& K(\hat{u}_{2\,\mathrm{H}}(t) - \hat{u}_{1\,\mathrm{H}}(t)) + F(t).
\end{aligned} \tag{G.9}$$

We want to derive the equation of motion for the expectation value of $\hat{u}_{1\,\mathrm{H}}(t)$; to this end, we begin by calculating $\frac{d^2}{dt^2}\hat{u}_{1\,\mathrm{H}}(t)$ from equation (G.8):

$$\frac{d^2}{dt^2}\hat{u}_{1\,\mathrm{H}}(t) = \frac{1}{m}\frac{d}{dt}\hat{p}_{1\,\mathrm{H}}(t). \tag{G.10}$$

We substitute the result (G.9) into the equation (G.10):

$$\frac{d^2}{dt^2}\hat{u}_{1\,\mathrm{H}}(t) = \frac{K}{m}(\hat{u}_{2\,\mathrm{H}}(t) - \hat{u}_{1\,\mathrm{H}}(t)) + \frac{F(t)}{m}. \tag{G.11}$$

Let us introduce the expectation value of $\hat{u}_{i\,\mathrm{H}}(t)$ as

$$\langle \hat{u}_{i\,\mathrm{H}}(t)\rangle \equiv \langle \psi(u_1, u_2, ..., u_\infty, 0)|\hat{u}_{i\,\mathrm{H}}(t)|\psi(u_1, u_2, ..., u_\infty, 0)\rangle \ , \tag{G.12}$$

where $|\psi(u_1, u_2, ..., u_\infty, 0)\rangle$ is the state of our system at $t = 0$. We obtain the following equation:

$$\frac{d^2}{dt^2}\langle \hat{u}_{1\,\mathrm{H}}(t)\rangle = \frac{K}{m}(\langle \hat{u}_{2\,\mathrm{H}}(t)\rangle - \langle \hat{u}_{1\,\mathrm{H}}(t)\rangle) + \frac{F(t)}{m}. \tag{G.13}$$

The expectation value of the displacement operator of the first atom is mathematically written as the convolution form of the Green's function G with the external force as [3]:

$$\langle \hat{u}_{1\,\mathrm{H}}(t)\rangle = \int_0^t G(t-\tau)F(\tau)d\tau. \tag{G.14}$$

The Laplace transformation of equation (G.14) is:

$$\langle \hat{u}_{1\,\mathrm{H}}(z)\rangle = G(z)F(z)\,, \tag{G.15}$$

where z is a coordinate in the complex space.

Proof. We suppose that

$$G(t-\tau) = 0 \quad \text{if} \quad t-\tau < 0. \tag{G.16}$$

$$\begin{aligned}\langle \hat{u}_{1\,\mathrm{H}}(z)\rangle &= \int_0^\infty \left[e^{-zt}\int_0^t G(t-\tau)F(\tau)d\tau\right]dt \\ &= \int_0^\infty \left[\int_0^\infty G(t-\tau)F(\tau)e^{-zt}d\tau\right]dt\,,\end{aligned}$$

where we used hypothesis (G.16). Now let us change the order of integration and call $t - \tau = t'$:

$$\begin{aligned}\langle \hat{u}_{1\,\mathrm{H}}(z)\rangle &= \int_0^\infty d\tau F(\tau)\int_0^\infty dt G(t-\tau)e^{-zt} \\ &= \int_0^\infty d\tau F(\tau)\int_{-\tau}^\infty dt' G(t')e^{-z(t'+\tau)}\,.\end{aligned}$$

Applying hypothesis (G.16) to the second integral to the right of the equal sign, we can write:

$$\begin{aligned}\langle \hat{u}_{1\,\mathrm{H}}(z)\rangle &= \int_0^\infty d\tau F(\tau) \int_0^\infty dt' G(t') e^{-z(t'+\tau)} \\ &= \int_0^\infty d\tau F(\tau) e^{-z\tau} \int_0^\infty dt' G(t') e^{-zt'} \\ &= F(z)G(z)\,.\end{aligned}$$

□

Our purpose is to derive a Green's function of a one-dimensional semi-infinite chain.

Let us start by adding an atom, labeled by $i = 0$, on top of the edge atom $i = 1$ of the one-dimensional chain. The new Hamiltonian system can be written as:

$$\hat{H} = \sum_{i=0}^{\infty} \frac{\hat{p}_i^2}{2m} + \sum_{i=0}^{\infty} \frac{1}{2} K(\hat{u}_{i+1} - \hat{u}_i)^2 - F_0(t)\,\hat{u}_0\,, \tag{G.17}$$

where $\hat{u}_0$ is the displacement operator of the new edge atom $i = 0$ and $F_0(t)$ is an external force applied only on the zero atom.

As before, we want to derive the equation of motion for the expectation value of $\hat{u}_{0\,\mathrm{H}}(t)$; repeating the same calculations as in (G.8), (G.9), we obtain:

$$\frac{d}{dt}\hat{u}_{0\,\mathrm{H}}(t) = \frac{i}{\hbar}[\hat{H}(t), \hat{u}_{0\,\mathrm{H}}(t)] = \frac{1}{m}\hat{p}_{0\,\mathrm{H}}(t)\,, \tag{G.18}$$

$$\frac{d}{dt}\hat{p}_{0\,\mathrm{H}}(t) = \frac{i}{\hbar}[\hat{H}(t), \hat{p}_{0\,\mathrm{H}}(t)] = K(\hat{u}_{1\,\mathrm{H}}(t) - \hat{u}_{0\,\mathrm{H}}(t)) + F_0(t)\,. \tag{G.19}$$

Therefore:

$$\frac{d^2}{dt^2}\hat{u}_{0\,\mathrm{H}}(t) = \frac{K}{m}(\hat{u}_{1\,\mathrm{H}}(t) - \hat{u}_{0\,\mathrm{H}}(t)) + \frac{F_0(t)}{m}\,, \tag{G.20}$$

and the equation for the expectation value $\langle \hat{u}_{0\,\mathrm{H}}(t)\rangle$ results:

$$\frac{d^2}{dt^2}\langle \hat{u}_{0\,\mathrm{H}}(t)\rangle = \frac{K}{m}(\langle \hat{u}_{1\,\mathrm{H}}(t)\rangle - \langle \hat{u}_{0\,\mathrm{H}}(t)\rangle) + \frac{F_0(t)}{m}. \tag{G.21}$$

Using the Laplace transformation we can rewrite the equation (G.21):

$$z^2\langle \hat{u}_{0\,\mathrm{H}}(z)\rangle = \frac{K}{m}(\langle \hat{u}_{1\,\mathrm{H}}(z)\rangle - \langle \hat{u}_{0\,\mathrm{H}}(z)\rangle) + \frac{F_0(z)}{m}. \tag{G.22}$$

Now, we can interpret $K(\langle \hat{u}_{0\,\mathrm{H}}(z)\rangle - \langle \hat{u}_{1\,\mathrm{H}}(z)\rangle)$ as the external force applied to the first atom, so equation (G.15) becomes:

$$\begin{aligned}\langle \hat{u}_{1\,\mathrm{H}}(z)\rangle =& KG(z)(\langle \hat{u}_{0\,\mathrm{H}}(z)\rangle - \langle \hat{u}_{1\,\mathrm{H}}(z)\rangle) \\ =& KG(z)\langle \hat{u}_{0\,\mathrm{H}}(z)\rangle - KG(z)\langle \hat{u}_{1\,\mathrm{H}}(z)\rangle \\ =& \frac{KG(z)\langle \hat{u}_{0\,\mathrm{H}}(z)\rangle}{1 + KG(z)}\,.\end{aligned} \tag{G.23}$$

By inserting equation (G.23) into equation (G.22), we obtain:

$$
\begin{aligned}
z^2 \langle \hat{u}_{0\,\mathrm{H}}(z)\rangle =& \frac{K}{m}\left(\frac{KG(z)\langle \hat{u}_{0\,\mathrm{H}}(z)\rangle}{1+KG(z)} - \langle \hat{u}_{0\,\mathrm{H}}(z)\rangle\right) + \frac{F_0(z)}{m} \\
=& \frac{K}{m}\left(\frac{KG(z)\langle \hat{u}_{0\,\mathrm{H}}(z)\rangle - \langle \hat{u}_{0\,\mathrm{H}}(z)\rangle - KG(z)\langle \hat{u}_{0\,\mathrm{H}}(z)\rangle}{1+KG(z)}\right) + \frac{F_0(z)}{m} \\
=& -\frac{K}{m}\frac{\langle \hat{u}_{0\,\mathrm{H}}(z)\rangle}{1+KG(z)} + \frac{F_0(z)}{m}.
\end{aligned}
\tag{G.24}
$$

Therefore:

$$
\begin{aligned}
\left(mz^2 + \frac{K}{1+KG(z)}\right)\langle \hat{u}_{0\,\mathrm{H}}(z)\rangle &= F_0(z) \\
\langle \hat{u}_{0\,\mathrm{H}}(z)\rangle &= \frac{F_0(z)}{mz^2 + \frac{K}{1+KG(z)}}.
\end{aligned}
\tag{G.25}
$$

We note that equation (G.25) is equivalent to equation (G.15) because the addition of the $i = 0$ atom to a semi-infinite chain in this way does not essentially change the original system due to its infinity. We deduce:

$$
G(z) = \frac{1}{mz^2 + \frac{K}{1+KG(z)}} = \frac{1+KG(z)}{mz^2 + mz^2KG(z) + K}. \tag{G.26}
$$

Equation (G.26) is readily solved:

$$
\begin{aligned}
mz^2G(z) + mz^2KG^2(z) + KG(z) &= 1 + KG(z) \\
mz^2KG^2(z) + mz^2G(z) - 1 &= 0
\end{aligned}
\tag{G.27}
$$

$$
\begin{aligned}
G(z) &= \frac{-mz^2 + \sqrt{(mz^2)^2 + 4mz^2K}}{2mz^2K} \\
&= -\frac{1}{2K} + \frac{1}{2mz^2K}\sqrt{m^2z^4 + \frac{4m^2z^4K}{mz^2}} \\
&= -\frac{1}{2K} + \frac{1}{2mz^2K}mz^2\sqrt{1 + \frac{4K}{mz^2}} \\
&= \frac{1}{2K}\left(-1 + \sqrt{1 + \frac{4K}{mz^2}}\right).
\end{aligned}
\tag{G.28}
$$

To calculate the normal modes we need to assume that the expectation value of $\hat{u}_{1\,\mathrm{H}}(t)$, that we define $u_1(t)$ for brevity of notation, is defined as

$$
u_1(t) = \int_{-\infty}^{t} G(t-\tau)F_1(\tau)d\tau\,, \tag{G.29}
$$

where $F_1(\tau)$ is an external force applied at the first atom.

The Fourier transform for $u_1(t)$ is

$$\begin{aligned}
\mathscr{T}(u_1(t)) &= \int_{-\infty}^{+\infty} dte^{i\omega t}u_1(t) \\
&= \int_{-\infty}^{+\infty} dte^{i\omega t}\int_{-\infty}^{t} d\tau\, G(t-\tau)F_1(\tau) \\
&= \int_{-\infty}^{+\infty} dte^{i\omega t}\int_{-\infty}^{\infty} d\tau\, G(t-\tau)F_1(\tau) \\
&= \int_{-\infty}^{+\infty} d\tau\, F_1(\tau)\int_{-\infty}^{+\infty} dt\, e^{i\omega t}G(t-\tau) \quad \text{we call } t-\tau=t' \\
&= \int_{-\infty}^{+\infty} d\tau\, F_1(\tau)\int_{-\infty}^{+\infty} dt'\, e^{i\omega(t'+\tau)}G(t') \\
&= \int_{-\infty}^{+\infty} d\tau\, e^{i\omega\tau}F_1(\tau)\int_{0}^{\infty} dt'\, e^{i\omega t'}G(t') = G(-i\omega)\mathscr{T}(F_1(\omega))\,,
\end{aligned} \tag{G.30}$$

where we used the hypothesis (G.16), $G(-i\omega)$ is the Laplace transform of the Green's function for the complex value $z=-i\omega$ and $\mathscr{T}(F_1(\omega))$ is the Fourier transform of the external force. Applying the Fourier transform to the equation of motion for the 0-th particle (G.21) and using the result (G.30) we still get the same equation (G.28) for the Laplace transform of the Green's function for $z=-i\omega$.

To search the normal modes, we suppose to use the following ansatz for the expectation value of the displacement of the i-th particle:

$$u_i(t) = \langle \hat{u}_{i\,\mathrm{H}}(t)\rangle = \overline{u}_i\, e^{i\omega t}\,. \tag{G.31}$$

Inserting this ansatz into equation (G.21) and assuming zero external force, we obtain:

$$\begin{aligned}
-m\omega^2\overline{u}_0\, e^{i\omega t} &= -K(\overline{u}_0-\overline{u}_1)\, e^{i\omega t} \\
m\omega^2\overline{u}_0 &= K(\overline{u}_0-\overline{u}_1)\,.
\end{aligned} \tag{G.32}$$

The relation (G.31) can be substitute into definition (G.29), obtaining:

$$\overline{u}_i\, e^{i\omega t} = \int_{-\infty}^{t} d\tau\, G(t-\tau)K(\overline{u}_0-\overline{u}_1)e^{i\omega\tau}\,, \tag{G.33}$$

where we considered $F_1(\tau)=K(\overline{u}_0-\overline{u}_1)e^{i\omega\tau}$ the external force acting on the first particle. Calling $t-\tau=t'$, we calculate

$$\begin{aligned}
\overline{u}_i\, e^{i\omega t} &= \int_{0}^{\infty} dt'\, G(t')K(\overline{u}_0-\overline{u}_1)e^{i\omega t}e^{-i\omega t'} \\
\frac{\overline{u}_1}{K(\overline{u}_0-\overline{u}_1)} &= \int_{0}^{\infty} dt'\, G(t')e^{-i\omega t'} = G(-i\omega)\,.
\end{aligned} \tag{G.34}$$

We note that the final expression is the Laplace transform of the Green's function for the complex value $z = -i\omega$. Therefore, we can write an explicit expression for $\overline{u}_1$:

$$\overline{u}_1 = \frac{G(-i\omega)K\overline{u}_0}{1+KG(-i\omega)} = \overline{u}_0 \frac{\frac{1}{2}(-1+\sqrt{1-\frac{4K}{m\omega^2}})}{1+\frac{1}{2}(-1+\sqrt{1-\frac{4K}{m\omega^2}})}$$
$$= \overline{u}_0 \frac{\frac{1}{2}(-1+\sqrt{1-\frac{4K}{m\omega^2}})}{\frac{1}{2}(1+\sqrt{1-\frac{4K}{m\omega^2}})} = \overline{u}_0 \frac{-1+\sqrt{1-\frac{4K}{m\omega^2}}}{1+\sqrt{1-\frac{4K}{m\omega^2}}}\,. \tag{G.35}$$

Iterating this technique for each particle of the chain, we obtain:

$$\overline{u}_i = \overline{u}_0 \left(\frac{-1+\sqrt{1-\frac{4K}{m\omega^2}}}{1+\sqrt{1-\frac{4K}{m\omega^2}}} \right)^i . \tag{G.36}$$

If $1-\frac{4K}{m\omega^2} \geq 0$, that is $\omega \geq 2\sqrt{\frac{K}{m}} \equiv \omega_c$, the fraction $\frac{-1+\sqrt{1-\frac{4K}{m\omega^2}}}{1+\sqrt{1-\frac{4K}{m\omega^2}}}$ is real with its module

$$\left| \frac{-1+\sqrt{1-\frac{4K}{m\omega^2}}}{1+\sqrt{1-\frac{4K}{m\omega^2}}} \right| < 1\,. \tag{G.37}$$

Therefore, for $\omega \geq \omega_c$ the 1D-crystal shows surface phonons decaying exponentially inside it.

If $\omega < \omega_c$ the square root $\sqrt{1-\frac{4K}{m\omega^2}}$ gives an imaginary number:

$$\sqrt{1-\frac{4K}{m\omega^2}} \equiv i\alpha\,, \tag{G.38}$$

and

$$\left| \frac{-1+\sqrt{1-\frac{4K}{m\omega^2}}}{1+\sqrt{1-\frac{4K}{m\omega^2}}} \right| = \left| \frac{-1+i\alpha}{1+i\alpha} \right| = 1\,. \tag{G.39}$$

Therefore, for $\omega < \omega_c$ the system shows acoustic phonons propagating within the 1D-crystal.

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