Thermal forces from a microscopic perspective

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Thermal gradients lead to macroscopic fluid motion if a confining surface is present along the gradient. This fundamental nonequilibrium effect, known as thermo-osmosis, is held responsible for particle thermophoresis in colloidal suspensions. A unified approach for thermo-osmosis in liquids and in gases is still lacking. Linear Response Theory is generalised to inhomogeneous systems, leading to an exact microscopic theory for the thermo-osmotic flow showing that the effect originates from two independent physical mechanisms, playing different roles in the gas and liquid phases, reducing to known expressions in the appropriate limits.

When a uniform bulk fluid is placed in a thermal gradient, mechanical equilibrium quickly sets in via the force balance condition, implying constant pressure throughout the system. In the absence of external forces, the steady-state is characterised by a space dependent density profile and a constant heat flux not associated to mass current [1]. The action of a thermal gradient on a fluid then resembles the effect of a fictitious “thermal mass current” [1] which has been known to play also a dynamical role since the first studies in gases by Feddersen, Crookes, Reynolds and Maxwell [3]. The onset of a stationary fluid flow induced by temperature gradients (in the absence of symmetry breaking forces like gravity, when convection dominates) is named thermo-osmosis [4] and only occurs due to the presence of a confining surface parallel to the thermal gradient, as already pointed out both in gases [5, 7] and in liquids [8, 10]. Thermo-osmosis is believed to be the driving mechanism for thermophoresis, i.e. the motion of a colloidal particle in a solvent due to a temperature gradient [11, 13], where the slip of the fluid in the boundary layer close to the particle’s surface gives rise to momentum transfer and eventually to particle motion. Thermo-osmosis is therefore one of the most fundamental manifestations of thermal forces and its physical origin is deeply rooted in nonequilibrium statistical mechanics. At the same time it is of great interest for applications as a mechanism for governing particle’s motion at the nanoscale [14, 15].

A unified description of thermo-osmosis is still lacking: The phenomenon was theoretically investigated mainly in the gas phase, where the fluid moves from the cold to the hot side and the characteristic length scale is of the order of the molecular mean free path [10]. The kinetic theory of gases has been used in this framework since the seminal work by Maxwell [7], who showed that the thermal creep is due to the tangential stress exerted by the gas on the fixed confining surface in the direction opposite to the temperature gradient. Such a stress, however, requires some exchange of energy and tangential momentum in the wall-particle scattering process and therefore depends on the modelling of fluid-surface interactions. Thermo-osmosis in the liquid regime is considerably less studied, both theoretically [11, 17, 19] and experimentally [11, 20, 22]. In addition, as shown in the recent review [23], experiments often disagree even about the direction of the thermo-osmotic flow. Nonequilibrium irreversible thermodynamics, based on the concept of local thermal equilibrium, was first used by Derjaguin to relate the thermo-osmotic velocity in liquids to the change of the local enthalpy of the fluid near the confining surface [10, 24]. The physical origin of the fluid motion, according to this macroscopic approach, is then due to the modification in the local thermodynamic properties of the fluid induced by the presence of a wall, as already pointed out in Ref. [25, 26]. Clearly, in the rarefied limit, which Derjaguin does not consider, the argument must fail because the effects of the wall on the (local) equilibrium properties of the gas disappear at low density. Only recently numerical simulations directly tackled this subtle nonequilibrium problem in the liquid regime [27–32], but a clear numerical evidence of the correctness of the Derjaguin formula has not been established yet. Rather, in Ref. [29] it was pointed out that Derjaguin expression cannot be correct because neither the enthalpy density nor the tangential pressure close to a surface is well defined on microscopic grounds.

This unsatisfactory setting calls for a first principle approach to the phenomenon, able to quantitatively evaluate the extent of the thermo-osmotic slip in terms of well defined properties of the fluid which can be measured in experiments and calculated in numerical simulations. In this Letter we present a microscopic description of thermo-osmosis on the basis of statistical physics: Linear Response Theory generalised to inhomogeneous and anisotropic environments. In the case of an imposed uniform thermal gradient, the use of conservation laws allows to evaluate the velocity profile of the fluid and the thermo-osmotic slip in terms of both the static and the dynamic equilibrium properties of the fluid near the surface: Close similarities to both the kinetic theory results and the Derjaguin formula are recovered by retaining each of these terms, showing that the gas and liquid regimes are indeed governed by different physical mechanisms.

The Green-Kubo formalism for linear response theory [33, 34] was generalised by Mori to deal with the ther-
nal transport coefficients \(^{33, 36}\). The starting point, as in the non equilibrium thermodynamics framework, is the concept of Local Equilibrium (LE) mathematically defined by the many body distribution function

\[
F^{LE} = Q^{-1} e^{-\int dr \beta(r) \mathcal{E}(r)},
\]

where \(Q\) is the partition function and the local energy density \(\mathcal{E}(r)\) is expressed in terms of the conserved densities as:

\[
\mathcal{E}(r) = \mathcal{H}(r) - u(r) \cdot \mathbf{j}(r) - \mu(r) \rho(r).
\]

Here \(\beta(r)\), \(u(r)\) and \(\mu(r)\) are external fields governing the temperature profile, the fluid velocity and the local chemical potential respectively. \(\mathcal{H}(r)\) is the microscopic many body Hamiltonian density

\[
\mathcal{H}(r) = \sum_i \delta(q_i - r) \left[ \frac{\mu_i^2}{2m} + \frac{1}{2} \sum_{j(i)} v(|q_i - q_j|) + V(q_i) \right]
\]

which describes a system of interacting point particles of mass \(m\) confined by hard walls represented by the external potential \(V(r)\). The operators

\[
\begin{align*}
\dot{\rho}(r) &= m \sum_i \delta(q_i - r), \\
\dot{\mathbf{j}}^\alpha(r) &= \sum_i \delta(q_i - r) p_i^\alpha,
\end{align*}
\]

define the local mass and momentum densities, which, together with the Hamiltonian \(\mathcal{H}(r)\) introduced in Eq. (2), satisfy microscopic conservation equations of the general form

\[
\frac{dA(r)}{dt} + \partial_\gamma \dot{J}^\gamma_A(r) = 0,
\]

where \(A(r)\) is the conserved density whereas \(\dot{J}^\gamma_A(r)\) is the corresponding current operator. Here and in the following Greek indices represent spatial components of vectors and tensors and Einstein summation convention is understood. In our case \(\dot{J}^\gamma_A(r)\) represents the mass current \(j^\gamma_m(r)\), the momentum \(\dot{J}^\gamma_j(r)\) and the energy flux \(\dot{J}^\gamma_e(r)\)

\[
\langle \dot{j}^\alpha(r) \rangle = \langle j^\alpha(r) \rangle_{LE} + \int_0^\infty dt \int dr \left[ \langle \dot{j}^\alpha(r, t) \dot{J}^\gamma_e(r') \rangle_0 \partial_\gamma, \beta(r') - \langle \dot{j}^\alpha(r, t) \dot{J}^\gamma_j(r') \rangle_0 \partial_\gamma, \beta \mu(r') \rangle - \langle \dot{j}^\alpha(r, t) \dot{J}^\gamma_m(r') \rangle_0 \partial_\gamma, \beta u^\gamma(r') \right].
\]

The averages \(\langle \ldots \rangle_0\) have been evaluated by means of the underlying equilibrium distribution \(F^{eq}\), and, to linear order in the velocity field, the LE distribution \(\mathcal{F}(r)\) gives \(\langle \dot{j}^\alpha(r) \rangle_{LE} = \rho(r) u^\alpha(r)\). Equation (6) is the formal expression of the thermo-osmotic slip in the presence of a non-uniform temperature field. Notice that (6) also involves odd rank tensors, forbidden by space isotropy, because this general theoretical framework also applies for a fluid close to an external surface, say a hard wall, which breaks isotropy defining a preferred direction.
Similar formulas can be derived for the averages of other physical quantities. The LE average of the momentum flux operator $\hat{J}^{\alpha\gamma}(r)$ gives the pressure tensor at equilibrium evaluated at the local temperature and chemical potential, but may also include a non-vanishing off diagonal contribution, as detailed in the SM.

However, in any experiment the external fields $\beta(r), u(r)$ and $\mu(r)$ appearing in Eq. (6) cannot be fixed from the outset but are rather self-consistently determined by the system, while the experimental set-up just defines the appropriate boundary conditions. Only pressure, temperature and velocity at the boundaries are given, while the spatial variation of the same quantities throughout the sample follow from the conservation equations: In steady-state conditions the divergence of the average momentum density $\langle \rho \hat{J}^z(r, t) \rangle_0$ and the results differ from those reported here. The boundary condition should be modified accordingly. Furthermore, the conservation law for the normal transverse pressure $p^{xz}(z) = p^{y\gamma}(z) = \rho r(z)$, the momentum density $\langle \hat{J}^x(r, t) \rangle_0$ and the source term $S(z)$ can be written as the sum of two distinct contributions $S(z) = S_a(z) + S_d(z)$ depending on the static and dynamical equilibrium correlations respectively:

$$S_a(z) = \int_{h/2}^z \frac{dz'}{h} \frac{\partial p_r(z')}{\beta \partial \beta} |_p - \int_0^\infty dt \int_0^\infty dr' (x - x') (\langle \hat{J}^x(r, t) \rangle_0 \hat{H}(r', t)|_0 - \hat{H}(r', t)|_0)$$

$$S_d(z) = \int_0^\infty dt \int_0^\infty dr' (\langle \hat{J}^x(r, t) \rangle_0 \hat{Q}(r', t)|_0$$

where we have introduced the heat flux operator $\hat{Q}_d(r) = J_{\beta\gamma}(r) - h_m \hat{J}_d^{\alpha}(r)$ and the operator $\hat{D}(r) = h_m \hat{p}(r) - \hat{H}(r)$.

To proceed further, let us consider a simple “slab geometry” where the fluid is confined between two infinite hard walls placed at a distance $h$ along the $z$ direction. The equilibrium density profile $\rho(r)$ is $z$-dependent and the only non vanishing components of the equilibrium pressure tensor define the normal $p^{xz}(z) = p_N(z)$ and the transverse pressure $p^{x\gamma}(z) = p^{y\gamma}(z) = \rho r(z)$. Furthermore, the width $h$ is chosen sufficiently large to guarantee that the fluid in the central region can be considered to a good approximation unaffected by the presence of the walls (in practice a few molecular diameters are sufficient).

A solution to the continuity equations is given by constant values of $\partial_s \beta$ and $\partial_s \beta \mu$, while the velocity field $u(z)$ is directed along the $x$-axis. Under these assumptions and within this simple geometry the stationary continuity equations for the average mass density $\langle \hat{\rho}(r) \rangle_0$, the energy density $\langle \hat{H}(r) \rangle_0$ and the $y$-component of the average momentum density $\langle \hat{J}^y(r) \rangle_0$ are identically satisfied. Furthermore, the conservation law for the normal (z) component of the momentum density $\langle \hat{J}^z(r) \rangle_0$ gives rise to the well known hydrostatic equilibrium condition

$$\partial_s \langle \hat{J}_j^{\alpha x}(r) \rangle_0 = \partial_s p_N |_{\beta(x), \mu(x)} = 0,$$

where the normal pressure is evaluated at the local temperature and chemical potential. The only non trivial continuity equation comes from the conservation of the $x$-component of the momentum density, which must be solved imposing that no pressure gradient is present far from the walls (open channel). The latter condition implies that $\partial_s [\beta \mu]$ can be expressed in terms of the temperature gradient by $\partial_s [\beta \mu] = h_m \partial_s \beta$, where $h_m$ is the enthalpy per unit mass of the fluid in the bulk. The detailed derivation is discussed in the SM. Here we report the final integro-differential equation for the velocity profile:

$$\int_0^h dz' K(z, z') \partial_z u^z(z’) = \partial_z \beta S(z).$$

The kernel $K(z, z’)$ is related to the local viscosity of the fluid

$K(z, z’)$ is chosen sufficiently large to guarantee that the fluid in the central region can be considered to a good approximation unaffected by the presence of the walls (in practice a few molecular diameters are sufficient).

The solution of the set of equations provides an expression for the gradient of the velocity field $\partial_z u^x(z)$ independent on the particular definition of the fluxes in [14], because the continuity equations only involve divergences of the fluxes (see [37]). When the result is substituted into Eq. (6) the final formula for the mass current is found:

$$\langle \hat{J}^x(z) \rangle = \rho(z) u^x(z) + \int_0^\infty dt \int_0^\infty dr’ \left[ \langle \hat{J}^x(r, t) \hat{J}^{\alpha x}(r') \rangle_0 \partial_s \beta - \beta \langle \hat{J}^x(r, t) \hat{J}^{\alpha x}(r') \rangle_0 \partial_s u^x(z’) \right].$$

All the contributions appearing in Eq. (10) vanish in the bulk, showing that the physical origin of thermo-osmosis relies on the existence of a confining surface [40]. However, the mass flux is not fully determined by Eq. (10) because the velocity field (and not only its derivative) appears in the first term. To resolve this ambiguity we have to know the mass flux at a given height $z$. This further requirement is not a limitation of the theory but rather a consequence of the Galilean invariance of the problem which, in an experimental set-up, is broken by the presence of friction between the fluid and the wall [41]. Instead, in the simplified model considered here, the wall is represented by an external confining potential (a hard wall) which does not modify the tangential
(x) component of the particles’ momenta. Supplementing this solution by a suitable (for instance no-slip) boundary condition for the mass flux, Eq. (10) allows to evaluate the thermo-osmotic flow in slab geometry: We first have to solve Eq. 7 for \( u^x(z) \) and then substitute the result into Eq. (10).

The above analysis of a model of simple fluid close to a wall is exact, within Linear Response Theory, and shows that two distinct mechanisms give rise to thermo-osmosis, both related to interface physics: The presence of anisotropies in the pressure tensor close to the wall (see Eq. (8)) and the effect of a confining surface on the dynamical correlation functions (see Eq. (9)). We now consider two limiting situations where these terms play a very different role in order to clarify their relevance in providing the required thermal force.

In liquids we expect that the dynamical correlations can be estimated by their bulk value and the kernel \( K(z, z') \) is taken to be a short ranged function

\[
K(z, z') \sim \eta \delta(z - z'),
\]

where \( \eta \) is the bulk viscosity of the fluid. Under these assumptions only the local equilibrium terms survive and the thermo-osmotic velocity reduces to \( u^x(z) \) given, for \( z < h/2 \), by

\[
u^x(z) = -\frac{\eta}{\rho} \int_0^{h/2} dz' \text{Min}(z, z') \frac{\partial p_T(z')}{\partial T} \,
\]

which coincides with the solution of the linearised Navier-Stokes equation for an incompressible fluid in the presence of a gradient in the tangential pressure given by the LE expression \[12, 29\]. Moreover, this solution closely resembles the prediction of Derjaguin approximation \[24\], where the enthalpy difference enters the integral \[42\]. The temperature derivative of the pressure tensor has been recently evaluated by numerical simulations \[20, 29\] for a Lennard-Jones fluid. Use of the numerical results allows to estimate that the resulting thermo-osmotic velocity is opposite to the thermal gradient and of the order of few micrometer per second.

In the low density limit, where kinetic theories provide a quantitative interpretation of the phenomenon \[10\], our formalism should reproduce the known results. Taking the ideal gas limit, i.e. ignoring the interparticle interactions, and introducing a finite relaxation time \( \tau \), the gas remains homogeneous and isotropic also close to the surface implying that \( S_d(z) = 0 \). The dynamical source term \( S_d(z) \) can be estimated retaining only the kinetic contribution to the equilibrium average in \[9\] as

\[
\int_0^\tau dt \sum_i \left\langle \delta(r - r_i(t)) \frac{p_i^x p_i^x(t)}{m} \frac{p_i^x - mh_m}{2m} \right\rangle.
\]

In the ballistic motion of each particle and in the elastic scattering against the wall the \( x \)-component of the momentum and the energy are conserved. The isotropy of the Maxwell distribution then leads to a vanishing result for \( S_d(z) \) due to the \( p_i^x \rightarrow -p_i^x \) symmetry of the equilibrium momentum distribution. A non-zero value can only be obtained if, during the scattering at the surface, at least one of these conservation laws are violated. For instance, if the scattering occurs at a time \( t_s < \tau \) and due to the collision the particle completely loses the memory about the initial value of the \( x \)-component of the momentum, then the equilibrium average in \[13\] will vanish for \( t > t_s \). This means that the contribution to \( S_d(z) \) coming from the particles with initial momentum in the direction opposite to the surface (i.e. those not suffering a collision against the wall) is not compensated by that due to the particles impinging on the surface. Therefore the thermal creep in gases sets in only assuming that during the particle-surface scattering the momentum is not conserved \[7, 43\]. This conclusion directly comes from the general Mori formalism and, under the hypothesis introduced above, the thermo-osmotic velocity \( v_\infty \) far from the surface reads

\[
v_\infty = \frac{3 \eta}{4 \rho} \frac{\partial_T}{T} = \frac{3}{4} k_B T \frac{\eta}{\rho} \frac{\partial_T}{T},
\]

which coincides with kinetic theory results \[7, 43\], showing how the slip velocity grows at low pressure, as experimentally demonstrated \[16\].

In summary, our generalisation of the Linear Response Theory formalism to inhomogeneous systems, applied to a simple microscopic model of fluid close to a planar smooth wall, has provided the general, exact, expression allowing to evaluate the thermo-osmotic flow. The emerging picture turns out to be more complex than expected on the basis of the previously adopted theoretical approaches, making use of kinetic theories as regards low pressure and rarefied gases and macroscopic linear irreversible thermodynamics for the liquid phase. The resulting velocity profile of the fluid \[10\] is valid for all regimes and depends on both static and dynamical equilibrium properties of the system (see Eqs. (7) and (10)): These expressions will be useful in the interpretation of future experiments and numerical simulations in the whole phase diagram of a fluid. A preliminary comparison with the existing macroscopic approach by Derjaguin shows that it closely resembles one of the two contributions found in our general expression. The other, instead, allows to reproduce the known expressions of the kinetic theory of gases in the appropriate limits.

Although our result is expressed in terms of quantitites, like the tangential pressure near the wall and the heat flux, which are not uniquely defined on microscopic grounds, the combination of these terms (see for instance Eq. (8)) is indeed independent of the adopted choice, thereby solving the problem posed in Ref. \[29\].

Our method is general: The results presented in the present letter can be easily extended to a closed channel, where the relevant quantity is the pressure difference be-
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\[
\frac{h_T(z)}{T} = \left. \frac{dp_T(z)}{dT} \right|_{\beta \mu}
\]
which differs from Eq. (12) due to the fact that here $\beta \mu$ and $p$ is kept constant in the derivation.
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SUPPLEMENTARY MATERIAL - Thermal forces from a microscopic perspective

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MICROSCOPIC CONSERVATION LAWS

The purpose of this Section is to provide the local expression of the fluxes which fulfil the microscopic counterpart of the macroscopic conservation laws of mass, momentum and energy. Furthermore we briefly discuss the ambiguity in the definition of the fluxes arising in microscopic conservation laws.

The general form of a local conservation law reads

$$\frac{dA(r)}{dt} + \partial_n J^A_n(r) = 0, \quad (S\ 1)$$

where the “operator” $A(r)$ represents the local conserved quantity and $J^A_n(r)$ the corresponding current. In the main text we have already introduced the mass, momentum and energy density “operators”, which we recall for convenience here

$$\dot{\rho}(r) = m \sum_i \delta(q_i - r), \quad (S\ 2)$$

$$\dot{J}_\alpha^\alpha(r) = \sum_i \delta(q_i - r) p_\alpha^i, \quad (S\ 3)$$

$$\dot{H} = \sum_i \delta(q_i - r) \dot{h}_i = \sum_i \delta(q_i - r) \left(\frac{p_i^2}{2m} + \frac{1}{2} \sum_{j \neq i} v(|q_i - q_j|) + V(q_i)\right), \quad (S\ 4)$$

where $q_i$ and $p_i$ are the generalised coordinates of the particle labelled by the index $i$. We remark that, according to the definition (S 3), the interaction energy $v_{ij} = v(|q_i - q_j|)$ between two particles $i$ and $j$ (located at $q_i$ and $q_j$) is ascribed without justification half to particle $i$ and half to particle $j$. Another admissible definition of the local energy density could ascribe the whole interaction energy $v_{ij}$ to the point $(q_i + q_j)/2$. The apparent ambiguity in (S 4) is related to the non local nature of the interparticle interaction potential $v(r)$ [1] and disappears integrating over the volume of the system (see also Ref. [2])

$$\dot{H} = \int dr \, \dot{H}(r) = \sum_i \frac{p_i^2}{2m} + \frac{1}{2} \sum_{j \neq i} v(|q_i - q_j|) + \sum_i V(q_i). \quad (S\ 5)$$

The expression of the fluxes directly follows from the evaluation of the time derivative of the conserved density (S 1), which reduces to the action of the Liouvillian $\mathcal{L}$ (corresponding to the Hamiltonian (S 3)) on $\dot{A}(r)$

$$\frac{dA(r)}{dt} = -\mathcal{L} \dot{A}(r), \quad (S\ 6)$$

where the expression of $\mathcal{L}$ is reported in standard textbooks (see e.g. Ref. [3]) and for a Hamiltonian of the form (S 3) reads

$$\mathcal{L} = \mathcal{L}_K + \mathcal{L}_v + \mathcal{L}_V$$

$$= -\sum_i \frac{p_i}{m} \cdot \frac{\partial}{\partial q_i} + \frac{1}{2} \sum_{i \neq j} \frac{\partial v_{ij}}{\partial q_i} \cdot \left(\frac{\partial}{\partial p_i} - \frac{\partial}{\partial p_j}\right) + \sum_i \frac{\partial V(q_i)}{\partial q_i} \cdot \frac{\partial}{\partial p_i}. \quad (S\ 7)$$

As regards the mass density (S 2) we straightforwardly get

$$\frac{d\dot{\rho}(r)}{dt} = -\mathcal{L} \dot{\rho}(r) = \sum_i \frac{\partial}{\partial q_i} \delta(q_i - r) \cdot p_i$$

$$= -\partial_n \dot{J}^\rho_n(r) \quad (S\ 8)$$

\footnote{Note that here the dependence on the phase space variables is understood.}
from which

$$\hat{j}_p^\alpha(r) = \sum_i \delta(q_i - r)p_i^\alpha. \quad (S\,9)$$

Analogously, the rate of change of $\hat{j}^\alpha(r)$ according to (S 6) provides a local conservation law corresponding to the macroscopic momentum balance equation:

$$\frac{d\hat{j}^\alpha(r)}{dt} = -\mathcal{L}\hat{j}^\alpha(r) = -\partial_\beta \left[ \sum_i \frac{p_i^\alpha p_i^\beta}{m}\delta(q_i - r) \right] - \frac{\dot{\rho}(r)}{m}\partial_\alpha V(r) - \frac{1}{2} \sum_{i \neq l} \frac{\partial V}{\partial q_i} [\delta(q_i - r) - \delta(q_l - r)]. \quad (S\,10)$$

The last term in Eq. (S 10) can be written as the divergence of a second rank tensor by means of the distributional identity [4]

$$\delta(q_j - r) - \delta(q_i - r) = \int_{C_{i\to j}} dy^\gamma \frac{\partial}{\partial y^\gamma} \delta(y - r) = -\partial_\gamma \int_{C_{i\to j}} dy^\gamma \delta(y - r), \quad (S\,11)$$

where the integral is along any contour $C_{i\to j}$ from $q_i$ to $q_j$. Making use of this result in Eq. (S 10) we obtain the microscopic continuity equation for the momentum density $\hat{j}^\alpha(r)$

$$\frac{d\hat{j}^\alpha(r)}{dt} = -\partial_\beta \hat{j}^\alpha\beta(r) - \frac{\dot{\rho}(r)}{m}\partial_\alpha V(r), \quad (S\,12)$$

where we have defined the microscopic momentum current operator $\hat{j}^\alpha\beta(r)$ as

$$\hat{j}^\alpha\beta(r) = \sum_i \frac{p_i^\alpha p_i^\beta}{m}\delta(q_i - r) + \frac{1}{2} \sum_{i \neq l} \frac{\partial V}{\partial q_i} \int_{C_{i\to j}} dy^\gamma \delta(y - r) \quad (S\,13)$$

$$= \sum_i \frac{p_i^\alpha p_i^\beta}{m}\delta(q_i - r) - \frac{1}{2} \sum_{i \neq l} \frac{q_i^\alpha}{|q_i|} \left. \frac{\partial V}{\partial q_i} \right|_{q_i=|q_i|} \int_{C_{i\to j}} dy^\gamma \delta(y - r), \quad (S\,14)$$

with $q_{ij} = q_j - q_i$. Note that the last term in Eq. (S 12) acts as a source contribution when a space dependent external field $V(r)$ is present. The average value of the operator $\hat{j}^\alpha\beta(r)$ is the so called (local) pressure tensor

$$p^{\alpha\beta}(r) = \langle \hat{j}^\alpha\beta(r) \rangle. \quad (S\,15)$$

Equation (S 13) shows how the local momentum current, which enters the continuity equation for the momentum density (S 12), can not be defined without ambiguity because different contours in (S 13) lead to different expressions for $\hat{j}^\alpha\beta(r)$ and also for the pressure tensor. For a system of particles interacting through central pair-wise additive potentials the average of Eq. (S 14) reads [3]

$$p^{\alpha\beta}(r) = \frac{\rho(r) k_B T}{m} \delta^{\alpha\beta} - \frac{1}{2} \int dy^{\alpha\beta} \frac{dv(|y|)}{|y|} \int_{C_{0\to y}} ds^\beta \rho^{(2)}(r - s, r - s + y), \quad (S\,16)$$

where $\rho^{(2)}(r, r')$ is the two-particle density [3] and the line-integral is extended, without any loss in generality [4], from the origin $0$ to a given point $y$. Making use of this result it is straightforward to show that in the homogeneous (and isotropic) limit the ambiguity in the definition of the pressure tensor disappears. Indeed the two particle distribution function can be expressed in terms of the radial distribution function ($\rho^{(2)}(r, r') = (\rho/m)^2 g(|r - r'|)$) and finally Eq. (S 16) reduces to

$$p^{\alpha\beta}(r) = \rho \delta^{\alpha\beta} = \frac{\rho k_B T}{m} \delta^{\alpha\beta} - \frac{1}{2} \frac{\rho^2}{m^2} \int dr \frac{r^{\alpha\beta}}{|r|} \frac{dv(|r|)}{|r|} g(|r|) \quad (S\,17)$$

2 Some references adopt a slightly different notation, introducing the stress tensor $\hat{\sigma}^{\alpha\beta}$ defined as $\hat{\sigma}^{\alpha\beta}(r) = -\hat{j}^{\alpha\beta}(r)$.

3 See [3], Eq. (3.2).
which is the well known virial (or pressure) equation for a homogeneous and isotropic fluid at density $\rho$.\footnote{\textsuperscript{3}}

The non uniqueness of the local pressure tensor has been first recognised by Kirkwood in the fifties. He provided an expression for the configurational contribution to the stress tensor in a paper with Buff\footnote{\textsuperscript{4}} and a different one in another work with Irving\footnote{\textsuperscript{5}}. The first explicit description of the ambiguity can be found in the paper by Harasima\footnote{\textsuperscript{6}} whereas a rigorous and exhaustive study of the problem was given in the eighties by Schofield and Henderson\footnote{\textsuperscript{3}}. This lack of uniqueness in the definition of the pressure tensor has been recently recovered in two papers\footnote{\textsuperscript{3, 10}} dealing with thermo-osmosis. The authors try to discriminate between different expressions of the pressure tensor estimating the value of the thermo-osmotic flow resulting from (approximate) predictions which involve the knowledge of the pressure tensor itself. In the most recent paper\footnote{\textsuperscript{10}}, they compare these predictions with the (exact) results obtained through a clever non equilibrium molecular dynamics simulation and they conclude that both the virial and the Irving-Kirkwood expression do not accurately predict surface forces due to temperature gradients.

However, we remark that the infinite possible definitions of the pressure tensor are indeed equivalent, i.e. all the physical observables must be invariant with respect to different choices of the path $C_{ij}$\footnote{\textsuperscript{3}}. As regards an inhomogeneous fluid, the pressure tensor itself is not a well defined observable on a length scale shorter than the correlation length or the range of the interparticle potential\footnote{\textsuperscript{1}}. Qualitatively, we can try to understand this circumstance reflecting on the fact that it is not possible to identify the surface where the pressure is acting. Analogously, we can not define without ambiguities the surface which separate two different phases of the same fluid. On the other hand, the pressure exerted on a given region of fluid and the surface tension of an interface are well defined quantities and do not depend on the particular definition of the pressure tensor\footnote{\textsuperscript{3}}.

As regards approximate theories, such as the local equilibrium assumption or the approach originally put forward by Derjaguin and recently adopted in\footnote{\textsuperscript{3, 11}}, the invariance of the observables with respect different definitions of the pressure tensor is not guaranteed a priori. However, the slip velocity of a fluid subject to a temperature gradient is a genuine physical quantity, also from the microscopic viewpoint. Therefore an exact prediction of the thermo-osmotic slip must be invariant on the choice of the trajectory in\footnote{\textsuperscript{13}}. We conclude that both the local thermal equilibrium and the Derjaguin approach include some errors, because their expression are not endowed by this invariance.

Finally, we note that the virial pressure tensor\footnote{\textsuperscript{17}}, evaluated in\footnote{\textsuperscript{9, 10}} in order to obtain the thermo-osmotic prediction of the thermo-osmotic properties of the fluid, does not correspond to any choice in the path\footnote{\textsuperscript{13}} and does not fulfil the hydrostatic balance condition for inhomogeneous fluids. This expression is commonly adopted within continuum hydrodynamics, where the relevant quantities vary on a length scale much larger than the correlation length\footnote{\textsuperscript{4}}.

The microscopic conservation law for the energy density $\hat{H}(r)$ can be obtained through the same steps followed before in the case of the mass current:

$$\frac{d\hat{H}(r)}{dt} = -\mathcal{L} \hat{H}(r) = -\mathcal{L}_K \hat{H}(r) - \sum_i \delta(q_i - r) \left[ \mathcal{L}_v + \mathcal{L}_V \right] \frac{\dot{p}_i^2}{2m}. \tag{S 18}$$

After some algebra, the action of the Lioulliians on the Hamiltonian reads

$$\mathcal{L}_K \hat{H}(r) = \sum_i \frac{p_i^2}{m} \left[ h_i \partial_i \delta(q_i - r) - \frac{\partial V(q_i)}{\partial q_i^a} \right] - \frac{1}{2m} \sum_{i \neq j} \delta(q_i - r) \frac{\partial v_{ij}}{\partial q_i^a} (p_i^a - p_j^a) \left[ \mathcal{L}_v + \mathcal{L}_V \right] \frac{\dot{p}_i^2}{2m} = \frac{\dot{p}_i^2}{m} \sum_{j \neq i} \frac{\partial v_{ij}}{\partial q_i^a} + \frac{\partial V(q_i)}{\partial q_i^a}. \tag{S 19}$$

Making use of these results in Eq. (S 18), we get

$$\frac{d\hat{H}(r)}{dt} = -\partial_a \left[ \sum_i \delta(q_i - r) \frac{\dot{p}_i^a}{m} \right] - \frac{1}{2m} \sum_{i \neq j} \frac{\dot{p}_i^a}{m} \frac{\partial v_{ij}}{\partial q_i^a} [\delta(q_i - r) - \delta(q_j - r)]. \tag{S 19}$$

The identity (S 11) allows to write Eq. (S 19) in the form of a microscopic conservation law:

$$\frac{d\hat{H}(r)}{dt} = -\partial_a \hat{j}_\alpha(r). \tag{S 20}$$
where we have defined the energy current $\hat{J}_H^\alpha (r)$ as

$$\hat{J}_H^\alpha (r) = \sum_i p_i^\alpha \delta (q_i - r) \hat{h}_i + \frac{1}{2} \sum_i \sum_{j \neq i} p_i^\alpha \frac{\partial \hat{h}_i}{\partial q_j} \oint_{C_{i \rightarrow j}} dy \delta (y - r). \quad (S\ 21)$$

Here we stress that Eq. (S\ 21) is the microscopic energy flux according to the definition of the local energy density given in Eq. (S\ 4). Different microscopic forms of the local energy result in different expressions of $\hat{J}_H^\alpha (r)$. In addition to this, the same considerations stated above for the momentum current apply: The ambiguity in the definition of the heat flux is recovered in the freedom connected to the choice of the path. However the thermal transport coefficients turn out to be independent on the particular choice in Eqs (S\ 21) and (S\ 4) (see Ref. [2]).

**EVALUATION OF AVERAGES**

**Local Equilibrium averages**

In this Section we evaluate the averages according to the local equilibrium distribution

$$F^{LE} = Q^{-1} e^{- \int dr \beta(r) \hat{E}(r)}, \quad (S\ 22)$$

where $\beta(r)$ is a scalar field related to the local temperature,

$$Q = \text{Tr} \left\{ e^{- \int dr \beta(r) \hat{E}(r)} \right\}$$

is the partition function and the local internal energy operator, already defined in the main text, reads

$$\hat{E}(r) = \hat{H}(r) - \hat{j}(r) \cdot \hat{u}(r) - \mu(r) \hat{\rho}(r).$$

The local Hamiltonian, momentum and particle densities have been defined in Eq. (S\ 4), (S\ 3) and (S\ 2) respectively and $\hat{u}(r)$ and $\mu(r)$ are the vector and scalar fields related to the local velocity profile and chemical potential (per unit mass) of the fluid respectively.

The averages according to the LE distribution (S\ 22) can be evaluated within linear response theory as follows. The essential hypothesis is that the non-equilibrium state defined by the LE distribution is very close, or analogously is a small perturbation, of an equilibrium state. First of all we introduce the underlying (zero order) equilibrium distribution function

$$F^{eq} = Q_0^{-1} e^{- \beta(H - m\mu N)} \quad (S\ 23)$$

defined by uniform temperature $\beta$ and chemical potential (per unit mass) $\mu$ and vanishing velocity field $u(r) = 0$. Here $Q_0$ is the equilibrium grand canonical partition function. The fields characterising the out of equilibrium state can then be written in terms of small deviations from the constant values of the temperature, the chemical potential and the vanishing velocity:

$$\beta(r) = \beta + \delta \beta(r); \quad \mu(r) = \mu + \delta \mu(r); \quad u(r) = 0 + \delta u(r).$$

Following the method inspired by linear response theory, we expand the LE distribution (S\ 22) about the equilibrium distribution (S\ 23) to the first order in the deviations $\delta \beta(r)$, $\delta \mu(r)$ and $\delta u(r)$. Noticing that the deviations from the equilibrium underlying distribution arise both in the exponential and in the partition function of $F^{LE}$ we get

$$F^{LE} = \frac{e^{- \int dr \beta(r) \hat{E}(r)}}{Q} \simeq \frac{Q_0 F^{eq} (1 - \hat{C}_e)}{Q_0 (1 - C_\beta)} \simeq F^{eq} (1 - \hat{C}_e + C_\beta),$$

7 The symbol $\text{Tr}\{\ldots\}$ represents the trace over all the degrees of freedom.
where the linear corrections to the exponential and the partition function can be written as

\[
\hat{C}_e = \int dr \left\{ \delta \beta(r) \left[ \hat{H}(r) - \mu \hat{\rho}(r) \right] - \beta \left[ \hat{j}(r) \cdot \delta \mathbf{u}(r) + \delta \mu(r) \hat{\rho}(r) \right] \right\},
\]

\[
C_Q = \int dr \left\{ \delta \beta(r) \left[ \left\langle \hat{H}(r) \right\rangle_0 - \mu \left\langle \hat{\rho}(r) \right\rangle_0 \right] + \beta \delta \mu(r) \left\langle \hat{\rho}(r) \right\rangle_0 \right\}.
\]  
(S 24)

Note that the averages \( \left\langle \ldots \right\rangle_0 \) are evaluated using the equilibrium distribution \( (S 23) \) and the difference in the notation underlines that \( \hat{C}_e \) still depends on the phase space coordinates. The final expression for the LE distribution within the linear approximation reads

\[
F^{LE} = F^{eq} \left\{ 1 - \int dr \left\{ \delta \beta(r) \left[ \hat{H}(r) - \mu \hat{\rho}(r) \right] - \beta \left[ \hat{j}(r) \cdot \delta \mathbf{u}(r) + \delta \mu(r) \hat{\rho}(r) \right] \right\} + C_Q \right\}.
\]  
(S 25)

Finally, the local equilibrium average of a given observable \( \hat{A}(r) \) is given by

\[
\left\langle \hat{A}(r) \right\rangle_{LE} = \left\langle \hat{A}(r) \right\rangle_0 - \int dr' \left\{ \delta \beta(r') \left[ \left\langle \hat{A}(r') \right\rangle \hat{H}(r') \right\rangle_0 - \mu \left\langle \hat{A}(r') \hat{\rho}(r') \right\rangle_0 \right] - \beta \left[ \left\langle \hat{A}(r) \hat{j}^\alpha(r') \right\rangle 0 \delta u^\alpha(r') \right] + \delta \mu(r') \left\langle \hat{A}(r) \hat{\rho}(r') \right\rangle_0 \right\} + C_Q \left\langle \hat{A}(r) \right\rangle_0.
\]  
(S 26)

Then, the local equilibrium averages of the particle density, of the momentum density and of the energy current read

\[
\left\langle \hat{j}^\alpha(r) \right\rangle_{LE} = \beta \int dr' \left\langle \hat{j}^\alpha(r') \right\rangle_0 u^\gamma(r') = \left\langle \hat{\rho}(r) \right\rangle_0 u^\alpha(r)
\]

\[
\left\langle \hat{j}^\alpha \hat{H}(r) \right\rangle_{LE} = \beta \int dr' \left\langle \hat{j}^\alpha \hat{H}(r') \right\rangle_0 u^\gamma(r')
\]

\[
\left\langle \hat{\rho}(r) \right\rangle_{LE} = \left\langle \hat{\rho}(r) \right\rangle_0 - \int dr' \left\{ \delta \beta(r') \left[ \left\langle \hat{\rho}(r') \right\rangle \hat{H}(r') \right\rangle_0 - \mu \left\langle \hat{\rho}(r') \hat{\rho}(r') \right\rangle_0 \right] - \beta \delta \mu(r') \left\langle \hat{\rho}(r) \hat{\rho}(r') \right\rangle_0 \right\} + C_Q \left\langle \hat{\rho}(r) \right\rangle_0
\]

where the last equality shows that the local equilibrium average of the density can be evaluated in the same way as the equilibrium average \( (S 23) \), provided that the temperature and the chemical potential are fixed on their local value in \( r \), that is \( \beta(r) \) and \( \mu(r) \).

The local equilibrium average of the momentum current reads

\[
\left\langle \hat{j}^\alpha \hat{\rho}(r) \right\rangle_{LE} = \left\langle \hat{j}^\alpha \hat{\rho}(r) \right\rangle_0 - \int dr' \left\{ \delta \beta(r') \left[ \left\langle \hat{j}^\alpha \hat{\rho}(r') \right\rangle \hat{H}(r') \right\rangle_0 - \mu \left\langle \hat{j}^\alpha \hat{\rho}(r') \hat{\rho}(r') \right\rangle_0 \right] - \beta \delta \mu(r') \left\langle \hat{j}^\alpha \hat{\rho}(r) \hat{\rho}(r') \right\rangle_0 \right\} + C_Q \left\langle \hat{j}^\alpha \hat{\rho}(r) \right\rangle_0.
\]  
(S 28)

Note that this LE average \( (S 28) \) is different from zero also if \( \alpha \neq \beta \), because, due to the configurational contribution in \( (S 13) \), \( \hat{j}^\alpha \hat{\rho}(r) \) is not an odd operator with respect to the momenta. It follows that the momentum flux tensor, which is diagonal in equilibrium systems, can acquire off diagonal components when the state of the system is described by a LE distribution as \( (S 22) \). However, it can be useful to point out that the diagonal components of this tensor can be written making use of the shorthand notation introduced in Eq. \( (S 27) \) as

\[
\left\langle \hat{j}^\alpha \hat{\rho}(r) \right\rangle_{LE} = \left\langle \hat{j}^\alpha \hat{\rho}(r) \right\rangle_0 \left. \right|_{\beta(r), \mu(r)} = p^{\alpha\beta}(r) \left. \right|_{\beta(r), \mu(r)}
\]

where \( p^{\alpha\beta}(r) \) is the equilibrium pressure tensor.

**Dynamical corrections to the Local Equilibrium averages**

The LE distribution function \( (S 22) \) introduced above is not stationary under the action of the Liouvillian \( (\mathcal{L}F^{LE} \neq 0) \) and it can not be used to evaluate the averages in the stationary thermo-osmotic flow. Indeed, if
the external constraints\footnote{i.e. the temperature, the chemical potential and the velocity fields.} are kept fixed, the actual phase space distribution will evolve in time due to the ensuing dynamics towards a stationary (time independent) out of equilibrium distribution. In order to obtain such distribution we follow, with slight changes, the approach proposed by Mori \cite{11,12}. Let us assume that the system is described at \( t = 0 \) by a non-stationary LE state \( F(t = 0) = F_{LE} \) and that for \( t > 0 \) the distribution \( F(t) \) evolves spontaneously according to the Liouville equation

\[
\partial_tF(t) = \mathcal{L}F(t). \tag{S 29}
\]

The formal solution of (S 29) can be written in an integral form as

\[
F(t) = F(0) + \int_0^t dt' \frac{d}{dt}F(t')
= F_{LE} + \int_0^t dt' \mathcal{L} \Psi(t')F_{LE}
= F_{LE} + \int_0^t dt' \Psi(t')\left[ L F_{LE} \right], \tag{S 30}
\]

where \( \Psi(t) = \exp\{t \mathcal{L} \} \) is the time evolution operator associated to the Liouvillian \( \mathcal{L} \).

The explicit evaluation of the right hand side of (S 30) is straightforward and reads

\[
F(t) = F_{LE} - \int_0^t dt' \int dr \Psi(t') \left\{ F_{LE} \beta(r) \left[ \partial_\alpha \hat{J}_\mu^\alpha(r) - \partial_\alpha \hat{J}_\gamma^\alpha(r) u^\alpha(r) - \mu(r) \partial_\alpha \hat{J}_\rho^\alpha(r) \right] \right\}
\]

where the action of the Liouvillian on the collisional invariants has been obtained in Eqs (S 20), (S 12) and (S 8). Assuming that the perturbation on the system due to the fields \( \beta(r), u(r) \) and \( \mu(r) \) is small we can restrict to a linear response approach and the (time dependent) average of a local observable \( \hat{A}(r) \) reads

\[
\langle \hat{A}(r) \rangle_t = \langle \hat{A}(r) \rangle_{LE} - \int_0^t dt' \int dr' \text{Tr} \left\{ \hat{A}(r) \Psi(t) \left[ F_{LE} \beta(r') \left( \partial_\alpha \hat{J}_\mu^\alpha(r') - \partial_\alpha \hat{J}_\gamma^\alpha(r') u^\alpha(r') - \mu(r') \partial_\alpha \hat{J}_\rho^\alpha(r') \right) \right] \right\},
\]

where the symbol \( \partial' \) represents the derivative w.r.t. \( r' \). Integrating by parts and taking the limit \( t \to \infty \) we obtain

\[
\langle \hat{A}(r) \rangle = \langle \hat{A}(r) \rangle_{LE} + \int_0^\infty dt' \int dr' \left[ \langle \hat{A}(r,t') \hat{J}_\mu^\alpha(r') \rangle_0 \partial_\alpha \beta(r') - \beta \langle \hat{A}(r,t') \hat{J}_\gamma^\alpha(r') \rangle_0 \partial_\alpha u^\alpha(r') \right.
- \left. \langle \hat{A}(r,t') \hat{J}_\rho^\alpha(r') \rangle_0 \partial_\alpha \mu(r') \right],
\]

where have shifted the time dependence on the observable \( \hat{A}(r) \) performing the canonical transformation \( \Psi(-t') \) and we have retained only the linear contributions.

**CONSTRAINTS FOR THE EXTERNAL FIELDS**

**Stationary continuity equations**

The expressions given above allow to evaluate the average of the relevant observables for a system out of equilibrium. However, the time independent fields \( \beta(r), \mu(r) \) and \( u(r) \), which enter these expressions, have not been fixed yet, and can not be determined \textbf{a priori}. In order to obtain their value, we will impose the physical constraints characterising a stationary state, that is the continuity equations satisfied by the average local energy density \( \langle H(r) \rangle \), by the average local momentum density \( \langle \hat{j}^\gamma(r) \rangle \) and by the average local particle density \( \langle \hat{\rho}(r) \rangle \). In formulæ

\[
\partial_\gamma \langle \hat{j}^\gamma(r) \rangle = 0, \tag{S 31}
\]

\[
\partial_\gamma \langle \hat{J}_\mu^\gamma(r) \rangle = 0, \tag{S 32}
\]

\[
\partial_\gamma \langle \hat{J}_\rho^\gamma(r) \rangle = -\frac{\langle \hat{\rho}(r) \rangle}{m} \partial_\alpha V(r). \tag{S 33}
\]
The solution of this set of five independent differential equations formally provides the values of the gradients of the fields $\partial_\alpha \beta(r)$, $\partial_\alpha \mu(r)$ and $\partial_\alpha u^\gamma(r)$. Unfortunately, without further approximations the general solution of this system cannot be obtained in closed form. On the other hand, when the equations are specialised to some simple geometry, symmetry considerations allow to considerably simplify the problem.

**Basic assumptions**

In what follows we will restrict our results to the system shown in Fig. 1 consisting of a fluid which fills the region between two infinite parallel planar walls, placed at a fixed distance $h$ (the so called slit or slab geometry). Furthermore we impose that the walls behave as hard objects with respect to the fluid and the fluid is kept out of equilibrium applying a temperature difference in the $x$ direction. The temperature difference is set at infinity and is such that the gradient is small and finite.

On the basis of the simple geometry of the problem, we expect that the solutions of the system of continuity equations (S 31), (S 32) and (S 33) will show some additional properties. Here we assume these properties and then we will show that such a solution exist. The assumptions on the solutions are the following:

1. The gradient of the field $\beta(r)$ is uniform throughout the fluid and is set in the $x$ direction:
   \[ \nabla \beta(r) = (\partial_x \beta, 0, 0) \tag{S 34} \]
   where $\partial_x \beta$ is a constant;

2. The gradient of the field $\mu(r)$ is uniform throughout the fluid and is set in the $x$ direction:
   \[ \nabla \mu(r) = (\partial_x \mu, 0, 0); \]

3. The only non vanishing component of the velocity field is along the $x$ axis and only depends on the coordinate $z$ normal to the wall:
   \[ u(r) = (u^x(z), 0, 0). \tag{S 35} \]

In the following we insert the assumptions just introduced in the the conservation laws (S 31), (S 32) and (S 33).

**Mass and energy conservation laws**

Due to the symmetries of the system, the steady state conservation law for the mass density (S 31) and for the momentum density (S 32) are identically satisfied.

---

*Note that within linear response theory this assumption together with (S 34) implies that $\partial_x (\beta \mu)$ is constant and $\nabla [\beta \mu](r) = (\partial_x (\beta \mu), 0, 0).$*
The average value of the momentum density is expressed in terms of the static and dynamical equilibrium averages as

\[
\langle \hat{j}^a(r) \rangle = \langle \hat{\beta}(z) \rangle_0 u^z(z) \delta^{ax} + \int_0^\infty dt' \int dr' \left[ \langle \hat{j}^a(r,t') \hat{J}_{H}^x(r') \rangle_0 \partial_x \beta \right. \\
- \beta \langle \hat{j}^a(r,t') \hat{J}_{H}^x(r') \rangle_0 \partial_x u^x(z) - \left. \langle \hat{j}^a(r,t') \hat{J}_{H}^\mu(r') \rangle_0 \partial_x (\beta \mu) \right],
\]

where we only made use of the assumptions introduced above. Due to the symmetry of the equilibrium system, only the \(x\) component of the average momentum density is non-vanishing (\(\langle j^y(r) \rangle = \langle j^z(r) \rangle = 0\))

\[
\langle \hat{j}^x(r) \rangle = \langle \hat{\beta}(z) \rangle_0 u^x(z) + \int_0^\infty dt' \int dr' \left[ \langle \hat{j}^x(r,t') \hat{J}_{H}^x(r') \rangle_0 \partial_x \beta \right. \\
- \beta \langle \hat{j}^x(r,t') \hat{J}_{H}^x(r') \rangle_0 \partial_x u^x(z) - \left. \langle \hat{j}^x(r,t') \hat{J}_{H}^\mu(r') \rangle_0 \partial_x (\beta \mu) \right]
\]

and the stationarity condition for the mass density (S 31) reduces to

\[
0 = \partial_a \langle \hat{j}^a(r) \rangle = \partial_x \int_0^\infty dt' \int dr' \left[ \langle \hat{j}^x(r,t') \hat{J}_{H}^x(r') \rangle_0 \partial_x \beta - \beta \langle \hat{j}^x(r,t') \hat{J}_{H}^x(r') \rangle_0 \partial_x u^x(z) - \left. \langle \hat{j}^x(r,t') \hat{J}_{H}^\mu(r') \rangle_0 \partial_x (\beta \mu) \right].
\]

The two point correlation functions only depend on the difference \(x-y\). After the integration over \(r'\) the dependence on \(x\) is lost and the derivative w.r.t. \(x\) vanishes.

Analogous considerations apply for the continuity equation for \(\hat{H}(r)\). Only the \(x\) component of the flux is different from zero:

\[
\langle \hat{J}_{H}^x(r) \rangle = \int dr' \langle \hat{J}_{H}^x(r) \hat{J}_{H}^x(r') \rangle_0 u^x(z) + \int_0^\infty dt' \int dr' \left[ \langle \hat{J}_{H}^x(r,t') \hat{J}_{H}^x(r') \rangle_0 \partial_x \beta \right. \\
- \beta \langle \hat{J}_{H}^x(r,t') \hat{J}_{H}^x(r') \rangle_0 \partial_x u^x(z) - \left. \langle \hat{J}_{H}^x(r,t') \hat{J}_{H}^\mu(r') \rangle_0 \partial_x (\beta \mu) \right].
\]

As before, the continuity equation, which in the stationary limit reduces to the derivative w.r.t. \(x\) of \(\langle \hat{J}_{H}^x(r) \rangle\), is identically satisfied because the integral of the correlation functions does not depend on \(x\).

### Momentum conservation law

The stationarity condition (S 33) gives origin to three independent equations. The projection of the momentum conservation law along both the \(y\) and \(z\) directions is identically satisfied whereas the projection along \(x\) determines the gradient of the velocity profile.

The stationary conservation law of \(\langle \hat{j}^z(r) \rangle\) requires the evaluation of the following quantities:

\[
\langle \hat{J}_{J}^{az}(r) \rangle = \langle \hat{J}_{J}^{az}(r) \rangle_{LE} + \int_0^\infty dt' \int dr' \left[ \langle \hat{J}_{J}^{az}(r,t') \hat{J}_{H}^z(r') \rangle_0 \partial_x \beta - \beta \langle \hat{J}_{J}^{az}(r,t') \hat{J}_{H}^z(r') \rangle_0 \partial_x u^z(z') \right. \\
- \left. \langle \hat{J}_{J}^{az}(r,t') \hat{J}_{H}^\mu(r') \rangle_0 \partial_x (\beta \mu) \right]
\]

\[
= \langle \hat{J}_{J}^{az}(r) \rangle_{LE} + \delta^{az} \int_0^\infty dt' \int dr' \left[ \langle \hat{J}_{J}^{az}(r,t') \hat{J}_{H}^z(r') \rangle_0 \partial_x \beta - \beta \langle \hat{J}_{J}^{az}(r,t') \hat{J}_{H}^z(r') \rangle_0 \partial_x u^z(z') \right. \\
- \left. \langle \hat{J}_{J}^{az}(r,t') \hat{J}_{H}^\mu(r') \rangle_0 \partial_x (\beta \mu) \right]
\]  

(S 37)

\[\text{10 The averages are evaluated at equilibrium and the system is homogeneous along the coordinate } x.\]
where in the second line we exploited the symmetries of the underlying equilibrium system as done before. The local equilibrium contribution, evaluated in Eq. (S 28) for a system without particular symmetries, reduces to

\[
\langle \hat{J}_j^{xx}(r) \rangle_{\text{LE}} = \langle \hat{J}_j^{xx}(z) \rangle_0 - \partial_x \beta \int dr' \cdot \left[ \langle \hat{J}_j^{xx}(r) \hat{H}(r') \rangle_0 - \mu \langle \hat{J}_j^{xx}(r) \hat{\rho}(r') \rangle_0 \right] \\
+ \beta \partial_x \mu \int dr' \cdot \langle \hat{J}_j^{xx}(r) \hat{\rho}(r') \rangle_0 + C_\Omega \langle \hat{J}_j^{xx}(z) \rangle_0.
\]  

(S 38)

Due to the translational symmetry, the term \( \langle \hat{J}_j^{xx}(r) \rangle_{\text{LE}} \) does not depend on \( y \) and does not contribute in (S 33). On the other hand, \( \langle \hat{J}_j^{xz}(r) \rangle_{\text{LE}} \) is different from zero due to the presence of the linear contributions in the derivatives of the fields in Eq. (S 38). These terms depend on \( z \), but do not depend on \( x \), because we can rearrange the integrals in (S 38) as

\[
\int dr' \cdot \langle \hat{J}_j^{xz}(r) \hat{O}(r') \rangle_0 = \int dr' \cdot \langle \hat{J}_j^{xz}(r) \hat{O}(r') \rangle_0 + x \int dr' \cdot \langle \hat{J}_j^{xz}(r) \hat{\rho}(r') \rangle_0 \\
= \int dr' \cdot \langle \hat{J}_j^{xz}(r) \hat{O}(r') \rangle_0,
\]  

(S 39)

where \( \hat{O}(r) \) is one of the scalar operator appearing in (S 38). Finally, the \( xz \) component of the LE pressure tensor reads

\[
\langle \hat{J}_j^{xz}(r) \rangle_{\text{LE}} = \int dr' \cdot \left[ \partial_x \beta \langle \hat{J}_j^{xz}(r) \hat{H}(r') \rangle_0 - \partial_x (\beta \mu) \langle \hat{J}_j^{xz}(r) \hat{\rho}(r') \rangle_0 \right],
\]  

(S 40)

where we remark that in the linear approach \( \partial_x (\beta \mu) = \mu \partial_x \beta + \beta \partial_x \mu \).

Finally, the LE average of the \( zz \) component of the momentum flux can be written as

\[
\langle \hat{J}_j^{zz}(r) \rangle_{\text{LE}} = p_N(z) \bigg|_{\beta(x), \mu(x)}.
\]

Note that \( \langle \hat{J}_j^{zz}(r) \rangle_{\text{LE}} \) depends both on \( z \) and \( x \), because the equilibrium averages are evaluated at the local value of the temperature and chemical potential \( \beta(x) \) and \( \mu(x) \).

Gathering the results obtained so far, the stationarity condition for the \( z \) component of the momentum density reads

\[
\partial_z \langle \hat{J}_j^{zz}(r) \rangle = \partial_z p_N(z) \bigg|_{\beta(x), \mu(x)} = -\frac{1}{m} \rho(z) \bigg|_{\beta(x), \mu(x)} \partial_z V(z),
\]  

(S 41)

where \( \rho(z) \) is the equilibrium density profile evaluated at the local \( \beta(x) \) and \( \mu(x) \), whereas for hard walls \( V(r) \) vanishes in the region occupied by the fluid. Eq. (S 41), the so called hydrostatic equilibrium condition, is always fulfilled by the normal component of the pressure tensor at each value of the coordinate \( x \) and is not specific to our problem. For hard walls (S 41) implies that the normal pressure is constant along \( z \) and equals the bulk pressure \( p \) at the local \( \beta(x) \) and \( \mu(x) \).

The stationarity condition for \( \langle j^y(r) \rangle \) is identically satisfied because the symmetry of the problem implies \( \partial_y \langle \hat{J}_j^{yy}(r) \rangle = 0 \).

The only non trivial continuity equation comes from the conservation of the \( x \) component of the momentum density

\[
\partial_t \langle \hat{J}_j^{xx}(r) \rangle = \partial_x \langle \hat{J}_j^{xx}(r) \rangle + \partial_x \langle \hat{J}_j^{xz}(r) \rangle = 0.
\]

It is straightforward to show that the relevant terms in this relation can be written as

\[
\langle \hat{J}_j^{xx}(r) \rangle = \langle \hat{J}_j^{xx}(r) \rangle_{\text{LE}} + \delta^{\alpha x} \int_0^\infty dt' \int dr' \left[ \langle \hat{J}_j^{xz}(r, t') \hat{J}_j^{xx}(r') \rangle_0 \partial_x \beta - \beta \langle \hat{J}_j^{xz}(r, t') \hat{J}_j^{xz}(r') \rangle_0 \partial_x u^x(z') \\
- \langle \hat{J}_j^{z}(r, t') \hat{\rho}(r') \rangle_0 \partial_x (\beta \mu) \right],
\]

where the tangential pressure acquires a dependence on \( x \) and reads

\[
\langle \hat{J}_j^{z}(r) \rangle_{\text{LE}} = p_T(z) \bigg|_{\beta(x), \mu(x)}.
\]
and $\langle \hat{J}_j^{zz}(r) \rangle_{L_K}$ has already been evaluated in Eq. (7).

Finally, the stationarity condition for the $x$ component of the momentum density reads

$$0 = \partial_x p_T(z)\bigg|_{\beta(x), \mu(x)} + \partial_z \int \text{d}r' (x - x') \left[ \partial_z \beta \left( \hat{J}_j^{zz}(r) \hat{H}(r') \right)_0 - \partial_x (\beta \mu) \left( \hat{J}_j^{zz}(r) \hat{\rho}(r') \right)_0 \right]$$

$$+ \partial_z \int_0^\infty \text{d}t' \int \text{d}r' \left[ \left( \hat{J}_j^{zz}(r, t') \hat{J}_j^{zz}(r') \right)_0 \partial_z \beta - \left( \hat{J}_j^{zz}(r, t') \hat{J}_j^{zz}(r') \right)_0 \partial_x (\beta \mu) \right]$$

$$- \beta \left( \hat{J}_j^{zz}(r, t') \hat{J}_j^{zz}(r') \right)_0 \partial_x u^x(z').$$ (S 42)

This condition is an integral equation for the velocity profile $u^x(z)$.

**Open channel**

The continuity equation for the momentum density along the $x$ direction (S 42) can be integrated provided we restrict to the free flow in an infinitely long channel (see Fig. 1), large enough to guarantee that in the central region the perturbation due to the walls is absent. In this region the fluid can be considered homogeneous and isotropic, and the normal and tangential components of the pressure tensor coincide and reduce to the bulk pressure $p$ of the fluid evaluated at the given value of the fields $\beta(x)$ and $\mu(x)$.

$$p_N(z \sim h/2)\bigg|_{\beta(x), \mu(x)} = p_T(z \sim h/2)\bigg|_{\beta(x), \mu(x)} = p \bigg|_{\beta(x), \mu(x)}.$$

Note that in the presence of a temperature gradient the bulk pressure itself acquires a dependence on the $x$ coordinate.

The free flow of fluid in an open channel imposes equal bulk pressure at the left and the right boundaries. Imposing this property, we find that the temperature and the chemical potential gradients will adapt in order to guarantee that

$$0 = \partial_x p \bigg|_{\beta(x), \mu(x)} = \partial_x \beta \left[ \partial_\beta p + \frac{\partial_\beta \mu}{\partial_\beta \mu} \partial_\mu p \right].$$

This equation determines the ratio between the temperature and the chemical potential gradients, which can be expressed in terms of thermodynamic densities as

$$\frac{\partial_x \mu}{\partial_x \beta} = -\frac{\partial_\beta p}{\partial_\mu p} = k_B T^2 \frac{s}{\rho},$$

where $s$ is the entropy density and we made use of the thermodynamic relations

$$\partial_T p \big|_\mu = -\partial_T (\Omega/V) \big|_\mu = s$$

$$\partial_\mu p \big|_T = -\partial_\mu (\Omega/V) \big|_T = \rho$$

where the derivatives of the grand potential $\Omega$ are always evaluated at constant volume $V$. The condition obtained above allows to express $\partial_x (\beta \mu)$ more naturally as

$$\partial_x (\beta \mu) = \partial_x \beta \left( \mu + T \frac{s}{\rho} \right) = \partial_x \beta \left( \frac{u + p}{\rho} \right) = \partial_x \beta h_m,$$

where $u$ is the internal energy and $h_m$ is the enthalpy per unit mass.

The stationarity condition (S 42) can be straightforwardly integrated along $z$ from 0 to $h/2$, where $h$ is the distance between the walls. All the integrals derived with respect to $z$ in (S 42) are odd functions of $z$ with respect to $h/2$ and, if evaluated at this point, vanish. The result of the integration turns out to be Equation (7) of the main text, that we report here for future reference:

$$\int_0^h \text{d}z' K(z, z') \partial_z' u^x(z') = \partial_x \beta S(z),$$ (S 43)

where the kernel $K(z, z')$ reads

$$K(z, z') = \beta \int_0^\infty \text{d}t' \int \text{d}r'_\perp \left( \hat{J}_j^{zz}(r, t') \hat{J}_j^{zz}(r') \right)_0.$$
and the source term $S(z)$ can be expressed as the sum of a static and dynamical contribution $S(z) = S_s(z) + S_d(z)$

$$S_s(z) = \int_{\mathcal{K}/2} d^2 x \left. \frac{\partial p_T(z')}{\partial \beta} \right|_{\beta} + \int d\mathcal{r}' (x - x') \langle \hat{J}^{\alpha}_j (r) \delta \hat{\mu}(r') \rangle_0,$$  \hfill (S 44)

$$S_d(z) = \int_0^\infty dt \int d\mathcal{r}' \langle \hat{J}^{\alpha}_j (r, t') \hat{J}_q(r') \rangle_0.$$  \hfill (S 45)

Note that the static contribution has been rewritten regarding the tangential pressure as a function of the $\beta(x)$ and of the bulk pressure $p$. Indeed, at midpoint

$$\left. p_T(h/2) \right|_{\beta(x), \mu(x)} \sim p$$

for each value of the coordinate $x$ and we can replace the local chemical potential with the bulk pressure obtaining

$$\partial_x p_T \bigg|_{\beta(x), p} = \partial_\beta p_T \partial_\beta \beta.$$  

**THERMO-OSMOSIS IN GASES**

In the ideal gas limit, i.e. ignoring the interparticle interactions, the momentum (S 13) and the energy (S 21) fluxes reduce to

$$\hat{J}^{\alpha}_j (r) = \sum_i \frac{p_i^\alpha p_i^\beta}{m} \delta(q_i - r); \quad \hat{J}_q^\alpha (r) = \sum_i \frac{p_i^2}{2m} \delta(q_i - r).$$

The goal of this section is to evaluate the mass current induced by the thermal gradient (S 36). We begin determining the solution for the velocity profile $u^x(z)$ and then we evaluate the dynamical contributions in (S 36).

**Solution of the equation for $u^x(z)$**

The static source term $S_s(z)$ vanishes because $p_T = p_N = p$ and the equilibrium average in (S 44) is performed on a quantity which is odd in the momenta. Then, the source term reduces to

$$S(z) = \sum_{i,j} \left\langle \int_0^\infty dt \int d\mathcal{r}' \delta (r - q_i(t)) \delta (r' - q_j(t)) \frac{p_i^\alpha(t) p_j^\beta(t)}{m} \frac{p_i^2}{2m} \left[ \frac{p_j^2}{2m} - mh_m \right] \right\rangle_0,$$  \hfill (S 46)

where the equilibrium average is evaluated according to the equilibrium distribution (S 28) and $q_i$ and $p_i$ are the coordinate and the momentum of the particle at $t = 0$ respectively. Without any kind of interaction between particles, the time integral in (S 46) is diverging because the correlations persist at all times. In order to mimic the behaviour of an almost ideal gas, where some collisions appear, we introduce a finite relaxation time $\tau$. This procedure allows to insert the collisions between ideal particles *a posteriori*, and $\tau$ turns out to be the mean time between two collisions of a given particle. In addition, only the contribution arising from the same particle (i.e. $i = l$) is non vanishing and the source term reads

$$S(z) = \sum_{i,j} \left\langle \int_0^\tau dt \int d\mathcal{r}' \delta (r - q_i(t)) \delta (r' - q_j(t)) \frac{p_i^\alpha(t) p_j^\beta(t)}{m} \frac{p_i^2}{2m} \left[ \frac{p_j^2}{2m} - mh_m \right] \right\rangle_0.$$  \hfill (S 47)

In the case of a perfectly reflecting wall, it is straightforward to show that the source term is zero. Indeed, specular reflections without energy exchange conserve both the $x$ component of the momentum and the modulus of the momentum. Then all the integrated quantities in Eq. (S 47) can be evaluated at time $t$. If we perform the canonical transformation $\mathcal{U}(-t)$ the average over the momenta does not depend on time, and the source term vanishes.

In agreement with the results obtained within kinetic theory [13], the occurrence of thermal creep is possible only assuming that in the particle-surface scattering the momentum is not conserved[11]. In order to mimic this behaviour

11 The additional contributions in Eq. (S 36) vanish for perfectly reflecting walls. See below.
and to obtain analytical results we assume that, due to the interaction with the surface during the scattering process, the \( x \) component of the particle’s momenta before and after the collision are fully uncorrelated. Furthermore, we restrict to the limiting semi-infinite geometry, where only the wall at \( h = 0 \) in Fig. 1 is present, in order to avoid multiple collisions between the surfaces.

The averages can be evaluated without any loss in generality within the canonical \((N, V, T)\) ensemble and the source term reads

\[
S(z) = \frac{N}{\bar{Q}_c} \int_0^\tau dt \int dr' \int dq \int dp \, \delta(r - q(t)) \, \delta(r' - q) \frac{p^x(t) p^x(t')}{m^2} \left[ \frac{p^2}{2m} - m\hbar \right] e^{-\beta p^2/2m},
\]

where \( \bar{Q}_c = V(2\pi m k_B T)^{3/2} \) and the factor \( N \) takes into account that the contributions in (S 47) arising from different particles are equal.

In order to evaluate the source term, let us briefly examine the behaviour of a particle before a given time \( t \) and for a given initial coordinate \( q \) as a function of the initial momentum \( p \). If \( p^x \geq -mq^x/t \), the particle does not bounce on the wall in the time interval \([0, t]\) and we can write

\[
p(t) = p; \quad q(t) = q + \frac{p}{m} t.
\]

On the other hand, when \( p^x < -mq^x/t \) the particle hits the wall at time \( t_s \), before \( t \). During the scattering the particle has completely lost the memory of the value of \( p^x \) before the bounce, therefore its self correlation is equal to 0 and the contribution in (S 48) arising from \( p^x < -mq^x/t \) vanishes. Therefore we can restrict the integral over \( p^x \) to the set \([-m q^x/t, +\infty]\) and, according to Eq. (S 49) we can write

\[
S(z) = \frac{N}{\bar{Q}_c} \int_0^\tau dt \int dr' \int dq \int dp_{\perp} \int_{-mq^x/t}^{+\infty} dp^x \, \delta\left(r - q - \frac{p}{m} t \right) \, \delta\left(r' - q\right) \frac{(p^x)^2 p^x}{m^2} \left[ \frac{p^2}{2m} - m\hbar \right] e^{-\beta p^2/2m},
\]

where the integral over the momentum \( p_{\perp} \) orthogonal to \( p^x \) is extended to \( \mathbb{R}^2 \). The final result for the source term, after a careful evaluation of the remaining integrals, reads

\[
S(z) = -\frac{N\pi m \tau}{\bar{Q}_c \beta^2} \exp \left[ -\beta \frac{m z^2}{2\tau^2} \right].
\]

Similar arguments allow to express the kernel as

\[
K(z, z') = \frac{N2\pi m^2}{\bar{Q}_c \beta^2} \Theta(z) \Theta(z') \exp \left[ -\beta \frac{m (z - z')^2}{2\tau^2} \right].
\]

Performing an appropriate change of variables, the differential equation (S 43) for \( u^x(z) \) can be written as

\[
\int_0^{+\infty} dz' \partial_z u^x(z') \frac{2m}{\tau k_B \Theta(x) T} e^{-z'^2 + \zeta z'} = 1,
\]

where \( \zeta = z \sqrt{m\beta/2\tau^2} \). The solution can be determined up to an additive constant \( C \) and reads

\[
u^x(z) = \frac{k_B}{2m} \tau \partial_x T \Theta(z + \delta) + C,
\]

where \( \delta \to 0^+ \) and the constant can be fixed imposing a given boundary condition. The relaxation time \( \tau \) introduced above can be related to the bulk viscosity \( \eta \), which appears in most of the results present in the literature [13, 14] and can be defined in terms of \( \tau \) as

\[
\eta = \beta \int_0^\tau dt \int dr' \langle \tilde{j}^x_z (r, t) \tilde{j}^x_z (r') \rangle_0.
\]

The integrals in (S 50) can be easily evaluated making use of the same arguments introduced above, and, after simple algebra, we obtain \( \eta = p \tau \). Finally, far from the wall the field \( u^x(z) \) can be written as

\[
u^x(z) = \frac{\eta}{p} \frac{k_B T}{2m} \partial_x T + C = \frac{\eta}{2\rho} \frac{\partial_x T}{T} + C.
\]
Mass current

As already stated, the velocity field does not have a direct physical meaning: The real flow is related to the average value of the $x$ component of the mass current (Eq. (10) of the main text), which we report here:

$$
\langle \hat{j}_x(z) \rangle = \rho u^x(z) + \int_0^T dt \int d\mathbf{r} \left\{ \langle \hat{j}_x(r,t) \rangle \left[ \tilde{J}_x^z(r') - h_m \hat{j}_x(r') \right] \right\}_0 \partial_x \beta - \beta \langle \hat{j}_x(r,t) \rangle_0 \partial_x u^x(z'),
$$

where $\rho(z)$ equals the bulk density for an ideal fluid and the flux $\tilde{J}_x^z$ has been written explicitly. It is straightforward to show that in the case of perfectly reflecting hard walls the mass current vanishes. As shown before, the velocity profile is equal to zero and for this reason the first and the last contribution in (S 52) vanishes whereas it is easy to prove that the other two contributions exactly cancel. Therefore, in order to obtain a net thermo-osmotic flow we must impose, as done before, a scattering process which induces an exchange of momentum between the particle and the wall. In doing so, the first contribution in (S 52) is trivial, whereas, after some algebra the integrals over $r'$ of the dynamical correlation functions read

$$
\int d\mathbf{r}' \langle \hat{j}_x(r,t) \rangle_0 \tilde{J}_x^z(r')_0 = \frac{5 N \pi m^2 2 \pi}{2 Q_c \beta} \frac{m \beta}{\beta} \left\{ \text{erf} \left( z \sqrt{\frac{\beta m}{2 t^2}} \right) + 1 \right\} - \frac{N \pi m^2 z}{Q_c \beta} \frac{1}{t} \exp \left( -\beta \frac{m z^2}{2 t^2} \right),
$$

$$
h_m \int d\mathbf{r}' \langle \hat{j}_x(r,t) \rangle_0 \tilde{j}_x^z(r')_0 = \frac{5 N \pi m^2 2 \pi}{2 Q_c \beta} \frac{m \beta}{\beta} \left\{ \text{erf} \left( z \sqrt{\frac{\beta m}{2 t^2}} \right) + 1 \right\},
$$

$$
\int d\mathbf{r}' \langle \hat{j}_x(r,t) \rangle_0 \partial_x u^x(z') = \frac{N m^2 \pi k_B \tau \partial_x T}{Q_c \beta t} z \exp \left( -\beta \frac{m z^2}{2 t^2} \right).
$$

The final result for the mass current, after the time integration is

$$
\langle \hat{j}_x(z) \rangle = \frac{\eta \partial_x T}{2 T} + \frac{\eta}{4} \left\{ \frac{2}{3 \pi} \frac{z}{\ell_g} \text{Ei} \left[ \frac{3}{2} \left( \frac{z}{\ell_g} \right)^2 \right] + \text{erf} \left( \sqrt{\frac{3}{2} \ell_g} \right) \right\} \partial_x T.
$$

where we have imposed no-slip boundary conditions for the mass current at $z = (12)$ and Ei($\cdot$) is the exponential integral and $\ell_g = \frac{\tau \sqrt{2/(m \beta)}}{}$. Far from the walls, for $z \gg \ell_g$ the exponential integral rapidly decays to 0 and the slip velocity $v_\infty = \langle \hat{j}_x(z) \rangle_{z \gg \ell_g}/\rho$ reduces to

$$
v_\infty \sim \frac{3 \eta}{4 \rho} \frac{\partial_x T}{T} = \frac{3}{4} k_B T \frac{\eta}{\rho} \frac{\partial_x T}{T}.
$$

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12 The constant turns out to be $C = \eta/4$. 

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It is clear that the constant $C$ in Eq. (10) of the main text is equal to $\eta/4$.