Convective heat transfer of a model emulsion at the droplet scale

Francesca Pelusi\textsuperscript{1}, Mauro Sbragaglia, and Roberto Benzi
Department of Physics, University of Rome “Tor Vergata” & INFN - Via della Ricerca Scientifica 1, 00133 Rome, Italy

Andrea Scagliarini and Massimo Bernaschi
Istituto per le Applicazioni del Calcolo, CNR - Via dei Taurini 19, 00185 Rome, Italy

Sauro Succi
Istituto per le Applicazioni del Calcolo, CNR - Via dei Taurini 19, 00185 Rome & Center for Life Nano Science Sapienza, Istituto Italiano di Tecnologia - Viale Regina Elena 295, I-00161 Rome, Italy

We numerically study the Rayleigh-Bénard (RB) convection in two-dimensional model emulsions confined between two parallel walls at fixed temperatures. The systems under study are heterogeneous, with finite-size droplets dispersed in a continuous phase. The droplet concentration is chosen to explore the convective heat transfer of both Newtonian (low droplet concentration) and non-Newtonian (high droplet concentration) emulsions, the latter exhibiting shear-thinning rheology, with a noticeable increase of viscosity at low shear rates. It is well known that the transition to convection of a homogeneous Newtonian system is accompanied by the onset of steady flow and a time-independent heat flux; in marked contrast, the heterogeneity of emulsions brings in an additional and previously unexplored phenomenology. As a matter of fact, when the droplet concentration increases, we observe that the heat transfer process is mediated by a non-steady flow, with neat heat-flux fluctuations, obeying a non-Gaussian statistics. The observed findings are ascribed to the emergence of space correlations among distant droplets, which we highlight via direct measurements of the droplets displacement and the characterisation of the associated correlation functions.

I. INTRODUCTION

Emulsions are complex and heterogeneous systems characterised by a collection of droplets of a dispersed phase in another continuous liquid phase. Emulsions are widely encountered in a variety of contexts, ranging from everyday life to high-tech applications \cite{1,2}. The dynamical behaviour of emulsions is intimately related to their rheological response, the latter encoded in the flow-curve of the material reporting the stress Σ as a function of the shear rate $\dot{\gamma}$, from which the effective viscosity $\eta_{\text{eff}}$ is extracted as $\eta_{\text{eff}} = d\Sigma/d\dot{\gamma}$. The rheology, in turn, depends on the droplet concentration: dilute emulsions behave as Newtonian fluids (i.e. $\eta_{\text{eff}}=\text{const}$) with a viscosity that increases with the droplet concentration \cite{6,15}. For larger concentrations, non-Newtonian effects emerge \cite{5,10}, the latter appear in the form of shear-thinning rheology, whereby the viscosity increases as the shear-rate decreases. This non-Newtonian behaviour is even more pronounced at larger droplet concentrations, where the emulsions can be categorised as yield stress materials \cite{17,19}, with a diverging viscosity at low $\dot{\gamma}$ (elastic behaviour), while exhibiting a finite viscosity at larger $\dot{\gamma}$. Studies on non-Newtonian emulsions – and more generally non-Newtonian complex fluids like those considered in this paper – typically refer to situations where the material response is analysed in the presence of external drivings, either a force or a shear. Further complications are brought about whenever mechanical solicitations arise from internal driving forces. A practical case in point is thermal convection \cite{21,22}, that we consider in this paper in the widely studied Rayleigh-Bénard (RB) set-up \cite{24,28}, consisting of a material between two walls heated from below and cooled from above (cfr. Fig. 1(a)). In this situation, the material is driven by buoyancy forces which depend on the local temperature field; the temperature field, in turn, is advected by the velocity field that diffuses in space via the viscosity of the material. For Newtonian fluids, an infinitesimal stress perturbation can linearly destabilise the conductive state if the advective time, which takes for a thermal perturbation (a “plume”) to travel from one wall to the other, is smaller than the time that it takes to be smeared out by thermal diffusion. There exists a critical ratio of these two timescales above which steady convection sets in \cite{29}. In the presence of non-Newtonian effects, this scenario is markedly modified, as witnessed by recent theoretical \cite{30,33}, numerical \cite{30,31,34} and experimental studies \cite{31,33,37}. Specifically, when the rheology changes from Newtonian to non-Newtonian, the stability of the base conductive state changes, to the point that for a yield stress material it becomes linearly stable \cite{29,30,31} and, for the onset of convection, a finite perturbation intensity is required; this finite perturbation value increases upon approaching the Newtonian critical point \cite{30}. Experimental studies show that the development of thermally induced perturbations is impacted by non-Newtonian properties as well \cite{31,33,36}. We note, however, that theoretical/numerical insights predominantly consider the problem of thermal convection in the presence of “local” rheology. In other words, it is assumed that the viscosity that enters the momentum
FIG. 1. Numerical simulations set-up. We study two-dimensional Rayleigh-Bénard (RB) convection of a model emulsion at changing droplets (dark-yellow domains) concentration while keeping fixed the temperature jump between the two walls. We consider both a Newtonian emulsion (NE) and a non-Newtonian emulsion (NNE) (panel (a)). The buoyancy forces are chosen to obtain the same time-averaged heat transport efficiency in both emulsions. Panel (b): rheological characterisation of both NE and NNE; in the inset, we show the effective dynamic viscosity $\eta_{eff} = d\Sigma/d\dot{\gamma}$ as a function of the stress $\Sigma$. The numerical simulations allow the Lagrangian tracking of the droplets (see box in panel (a)): starting from the Eulerian droplets displacement $d(x, y, t)$ (black arrows in panels (a) and (c)) we construct the associated fluctuations with respect to its time average $\delta d(x, y, t) = d(x, y, t) - \langle d(x, y, t) \rangle$; (panel (d), see text for more details). All dimensional quantities are reported in simulation units.

The equation depends locally on $\dot{\gamma}$. This assumption may be reasonable whenever convection is treated on "continuum scales", i.e. at scales much larger than the characteristic size of the constituents of the material. When we move to scales comparable with that of the constituents, it is known that a description based on a local relation between $\Sigma$ and $\dot{\gamma}$ falls short of capturing the relevant physics, and finite-size effects need to be taken into account to obtain a comprehensive characterisation of the flow [38–43]. This sets a compelling case for the study of thermal convection at those small scales. In the RB set-up under consideration, this can be accomplished by considering confined systems, with a wall-to-wall distance $H$ of the order of a few tens of constituents size (cfr. Fig. 1(a)). It was argued that convective transport of non-Newtonian complex fluids, might be impacted in a non-trivial way by "rearrangements" of the mesoscopic constituents at small scales, but unfortunately, due to the limited resolution, the available experimental data were not conclusive [35]. Here, we take a step further and provide a detailed characterisation of the heat transfer properties just above the transition from conduction to convection. Our work hinges on numerical simulations that allow an unprecedented detailed analysis of heat transfer, thereby permitting to highlight both the role of finite-size constituents and their space-time correlations. This paper is organised as follows: in Section II we report the essential features of the set-up used for the numerical simulations with the associated tools of analysis; in Section III we present and discuss the numerical results; conclusions will be drawn in Section IV.

II. METHODS

We report the results of numerical simulations of RB thermal convection in stabilised two-dimensional emulsion systems. The choice of the dimensionality is instrumental to properly resolve the emulsion droplets inside the simulation and achieve reasonable statistics within a few days of simulation time. As to the numerical technique, we resort to the mesoscale lattice Boltzmann method (LBM) [44, 45]. Briefly, the model allows the simulation of two-component fluids undergoing phase segregation, wherein the system can be divided into bulk regions with majority of one of the two components. Coalescence of bulk domains is further inhibited by the introduction of repulsive interface forces (disjoining pressure). Thus, the system can be prepared with a number of droplets $N_{\text{droplets}}$ (dispersed phase) inside the contin-
ous phase (cfr. Fig. 1(a)). The concentration of the droplets is a tunable parameter in the preparation of the system, thus we can explore situations ranging from dilute to denser concentrations. The emulsion is placed between two walls at a distance of $H$: the channel size is chosen to have the ratio $H/d \sim 25$, where $d$ is the mean droplet diameter. It is further subject to buoyancy forces. At hydrodynamical scales the reference dynamical equations are the diffuse-interface Navier-Stokes equation for the hydrodynamical field $\mathbf{u}(x, y, t) = (u_x, u_y)(x, y, t)$ (repeated indexes are summed upon)

$$\rho \left( \partial_t u_i + u_k \partial_k u_i \right) = -\partial_j \left[ -P_{ij} + \eta_0 \left( \partial_i u_j + \partial_j u_i \right) \right] + \rho \alpha g T \delta_{iy}, \quad i, y = x, y,$$

where $\rho$ is the local total density, $P_{ij}$ the non-ideal pressure tensor, $\eta_0$ the dynamic viscosity of the bulk phase, $\alpha$ the thermal expansion coefficient and $g$ the gravity acceleration. The temperature field $T(x, y, t)$ (taken as relative to some reference temperature) obeys the advection-diffusion equation

$$\partial_t T + u_k \partial_k T = \kappa \partial_{kk} T.$$

Table I. We report the buoyancy amplitudes necessary to obtain the same time-averaged Nusselt number $(\text{Nu})_t$ (see text for more details). All dimensional quantities are reported in simulation units.

| Type  | $(\text{Nu})_t$ | $\alpha g \Delta T$ |
|-------|----------------|---------------------|
| NNE   | 2.0            | $5.96 \times 10^{-6}$ |
| NE    | 2.0            | $6.65 \times 10^{-7}$ |
| NNE   | 2.7            | $8.71 \times 10^{-6}$ |
| NE    | 2.7            | $1.42 \times 10^{-6}$ |
| SP    | 2.7            | $3.51 \times 10^{-6}$ |

### III. RESULTS & DISCUSSION

We consider two distinct emulsions with different volume fractions: a dilute Newtonian emulsion (NE, hereafter) with $N_{\text{droplets}} = 700$ and a concentrated non-Newtonian emulsion (NNE, hereafter) with $N_{\text{droplets}} = 1000$; see Fig. 1(a) to get a pictorial view of how the system looks like and Fig. 1(b) for flow curves from shear rheology measurements on the two types of emulsion. The droplet concentration of the NNE is the largest one for which we do not observe coalescence events during the time dynamics, such that the number of droplets and the polydispersity are kept fixed. For the sake of a fair comparison between NNE and NE we have, therefore, decided to neglect larger concentrations. Notice that the droplet concentration for the NNE is large enough to detect an "incipient" yield stress behaviour. The buoyancy amplitude, $\alpha g \Delta T$, is chosen in such a way that the system sustains a convective state, just above the transition from conduction to convection. In order to address the heat transfer properties we focus on the dimensionless time-dependent Nusselt number [27, 49–51]:

$$\text{Nu}(t) = \frac{\left\langle u_y(x, y, t)T(x, y, t) \right\rangle_{x,y} - \kappa \left( \partial_y T(x, y, t) \right)_{x,y}}{\kappa \Delta T}$$

where $\left\langle \ldots \right\rangle_{x,y}$ stands for the spatial average. Nu is a parameter that quantifies the relative intensity between convective and conductive transport. The two emulsions have different effective viscosities, with NNE being more viscous than NE, hence they respond differently to a given imposed buoyancy amplitude. Specifically, in order to observe the same heat transport efficiency (i.e. same Nusselt number) in both systems, it is necessary to impose a larger buoyancy in the dynamical evolution (cfr. Eq. (4)) if the emulsion is more concentrated (non-Newtonian behaviour). Our initial strategy was to determine – for each emulsion – the buoyancy amplitude necessary to obtain the desired value of the time-averaged Nusselt number $\langle \text{Nu} \rangle_t$ (see Table I for details). In doing so, a first marked difference emerges in the comparison between NE and NNE. While NE can flow with a Nusselt number that is essentially independent of time, NNE shows neat and larger fluctuations in the Nusselt number (cfr. Fig. 3). To dig deeper into this phenomenol-

\[\text{This is the viscosity that the system would exhibit in the presence of a homogeneous continuous phase without droplets.}\]
ogy, we also report in Fig. 2(c) the 2D-maps of the local shear \( \dot{\gamma} \) in correspondence of a local maximum/minimum in \( \text{Nu}(t) \) for NNE. These maps clearly show the coexistence of spatial regions, of different extent, at very small (dark regions) and larger (light regions) shear rates \( \dot{\gamma} \), respectively. In other words, while in correspondence of a maximum in the Nusselt number, the system is predominantly fluidised with a little number of small shear rates (i.e. large viscosity) regions, in correspondence of a minimum in the Nusselt number, the reversed situation holds. We remark that for a Newtonian fluid at these values of the Nusselt number the convective states are time-independent, hence it is natural to ask where these fluctuations come from. To get further insight into the problem, we studied also the case of a single-phase (SP) fluid model with a “local” closure for the effective dynamic viscosity. More precisely, we fitted the rheological curve of NNE displayed in Fig. 1(b) and extracted the effective “local” viscosity from the slope, \( \eta_{\text{eff}}(\dot{\gamma}) = d\Sigma/d\dot{\gamma} \); we then ran a numerical simulation with Eqs. 1 with the so constructed \( \eta_{\text{eff}}(\dot{\gamma}) \), however without droplets. In this way, we are simulating a homogeneous fluid inheriting the complex rheology of the emulsion via a local relationship between the dynamic viscosity and the local shear rate. Also in this case, for an optimal comparison, a different buoyancy has been imposed, such as to keep the time-averaged Nusselt number \( \langle \text{Nu} \rangle_t \) fixed. Table I shows the numerical values of the buoyancy amplitudes \( \alpha \Delta T \) used in the simulations. With respect to the NNE, we remark that it is necessary to reduce the buoyancy amplitude \( \alpha \Delta T \) of about 60% in the SP case in order to obtain the same heat transfer of the NNE. In other words, the SP case does not reproduce the same time-averaged Nusselt number \( \langle \text{Nu} \rangle_t \) for the same buoyancy amplitude of the NNE. Moreover, as shown in Fig. 2(b), the fluid with the “local” rheology does not show any fluctuations of \( \text{Nu}(t) \). The conclusion that we draw is that the SP is unable to predict both the time-averaged Nusselt number as well as its fluctuations; moreover, the observed fluctuations emerge due to the simultaneous presence of non-Newtonian rheology and finite-size droplets. We deemed, therefore, appropriate to inspect this phenomenology from a Lagrangian viewpoint, that is looking at the relevant observables along a droplet trajectory; in particular, inspired by Lagrangian studies of turbulent RB convection [12, 53], we focus on the droplet Nusselt number \( \text{Nu}_{i \text{drop}}^{(\text{drop})} \). The definition of this Lagrangian observable is constructed in such a way that the global Nusselt number \( \text{Nu} \) may be seen as the sum over the local contributions of the single droplets, i.e.

\[
\text{Nu}(t) = \frac{1}{N_{\text{droplets}}} \sum_{i=1}^{N_{\text{droplets}}} \text{Nu}_{i \text{drop}}^{(\text{drop})}(t).
\]

A good candidate to satisfy Eq. 5 is the droplet Nusselt number defined as:

\[
\text{Nu}_{i \text{drop}}^{(\text{drop})}(t) = \frac{u_{g}^{(i)}(t)T^{(i)}(t) - \kappa (\partial_{y}T)^{(i)}(t)}{\kappa \frac{\Delta T}{H}}.
\]

where \( u_{g}^{(i)}(t) = u_{g}(X_{i}(t), t), T^{(i)}(t) = T(X_{i}(t), t) \) and \( (\partial_{y}T)^{(i)}(t) = \partial_{y}T(X_{i}(t), t) \) are the fluid velocity, temperature, and temperature gradient evaluated at the position of the \( i \)-th droplet centre-of-mass, \( X_{i}(t) \). We report the PDF of the droplet Nusselt number for the numerical simulations previously analysed in Fig. 2. For the sake of comparison, we show data with the \( x \)-axes given in units of the standard deviation with respect to the average value. Being the SP simulation without droplets, for the computation of \( \text{Nu}_{i \text{drop}}^{(\text{drop})} \) we took an Eulerian viewpoint and divided...
the thermal convection, hence providing enhanced posi-
tive tails in the PDF of the droplet Nusselt number (red box in the top panel of Fig. 3(b)); rearrangements may also inhibit convective transport, hence a contribution to the negative tail of the PDF (blue box in the bottom panel of Fig. 3(b)). Notice also that such “extreme” events are located within the boundary layers. Echoing the observations made for Fig. 3 we remark that the enhancement of the tails appears only in the presence of finite-size droplets, whereas the SP model does not show such pronounced tails, being closer to the NE case. The analysis performed in Fig. 3 helps in further elucidating the large scale fluctuations in the Nusselt number observed in Fig. 2. In particular, it gives some hints on the physical mechanism that allows the system to display the switch shown in Fig. 2(c). If the system is almost entirely non-fluidised, it can change to a situation where it is predominantly fluidised if non-local correlations are active into the system. The same holds true for a system that is predominantly fluidised and switches back to an almost entirely non-fluidised state. Spatially extended correlated zones are also expected by looking at the maps of $\tilde{d}(x,y,t)$ reported in Fig. 2(b), where collective “bursts” of $\tilde{d}(x,y,t)$ appear. In fact, in the absence of space correlations, “bursts” of activity would be unable to propagate in the system and trigger the switching of a substantially large part of the system in another state. These facts said it comes as a logical consequence to study the observable $\delta d(x,y,t)$ to better corroborate the existence of non-trivial correlations in the system. To this aim, we first average the field $\delta d(x,y,t)$ in the $x$-direction, i.e. $\delta d(y,t) = \langle \delta d(x,y,t) \rangle_x$. The space-time evolution of the displacement fluctuations is reported in Fig. 4(a), where we plot the absolute value of $\delta d(y,t)$ in the $(y,t)$ plane. It is seen that for the NNE the displacement fluctuations depart from zero coherently in extended space regions, predominantly close to the boundaries. Such space coherence persists for some finite time. This is in marked contrast with the observations for the NE, where the space-time coherence is visibly lost. Finally, to unveil more quantitatively the difference in space correlations between NE and NNE, we have computed the spatial correlation function $C(r)$. To this aim, we have adapted the definitions of previous literature studies \cite{54, 55, 56} to the absolute value of $\delta d(y,t)$:

$$C(r) = \frac{\langle |\delta d(0,t)| |\delta d(r,t)| \rangle_x - \langle |\delta d(0,t)| \rangle_x \langle |\delta d(r,t)| \rangle_x}{\langle |\delta d(0,t)| \rangle_x \langle |\delta d(r,t)| \rangle_x}$$

where $-H/2 < r < +H/2$ and $\langle |\delta d(0,t)| \rangle_x \langle |\delta d(r,t)| \rangle_x$ is the standard deviation of $|\delta d(0,t)|$ ($|\delta d(r,t)|$). In Fig. 4(b) we show $C(r)$ for both NE and NNE with the $x$-axis normalised by the mean droplet diameter $d$: while for the NE case the correlation rapidly decays to zero within a distance of the order of single droplet diameter, the NNE emulsion shows larger correlation extending in space for a markedly larger distance.

It is worth noting that two reference cases studied here are representative of two different “categories” of emul-
IV. CONCLUSIONS

We analysed the heat transfer properties of a model emulsion in the Rayleigh-Bénard (RB) set-up, where the emulsion is placed in a confined cell between two parallel walls, heated from below and cooled from above. The gap of the cell has been chosen to accommodate a few tens of droplet diameters. This is an optimal set-up to highlight the heat transfer properties at the droplets scales, i.e., at those scales where the size of the regions studied becomes of the order of the system constituents. The droplets concentration has been changed to provide a systematic comparison on the heat transfer properties between two representative emulsion concentrations: a Newtonian emulsion (NE) exhibiting a Newtonian rheology, and a non-Newtonian emulsion (NNE) exhibiting shear-thinning rheology with a marked increase of the viscosity at low shear rates. We focused our analysis just above the onset of convection, by monitoring the time behaviour of the Nusselt number $\text{Nu}(t)$, i.e., the ratio of convective to conductive heat transfer. It is observed that NE can sustain a steady convective heat transfer efficiency, as expected for a homogeneous Newtonian fluid just above the onset of convection; the phenomenology for the convective heat transport for NNE is manifestly different, and shows neat fluctuations in the Nusselt number, corresponding to the switching between two qualitatively different system configurations, with a predominance of fluidised (high Nusselt number) and non-fluidised (low Nusselt number) regions. This goes together with the emergence of fat tails in the statistics of the local Nusselt number, i.e., the Nusselt number at the droplet scale. Overall, the convective phenomenology for the NNE is attributed to the combined effect of non-Newtonian rheology and a finite correlation between distant droplets, which we have unveiled via the analysis of the displacement fields.

On a more general perspective, a few other remarks are in order. The droplet concentration of the analysed NNE is such that its rheology shows the germinal signatures of an incipient yield stress rheology; for the emulsion to be categorised clearly as a yield stress material [19], larger concentrations are needed. The convective states at those larger concentrations, however, could not be explored in detail in this paper because of the occurrence of droplets coalescence during motion that alters the system response. This calls for the development of improved numerical models, able to prevent the coalescence. Beyond the methodological motivation, we stress that such dedicated numerical studies would be physically relevant, as one may argue that the observed findings can be magnified in yield stress materials. In this scenario, our findings raise interesting questions as to the precise meaning of viscosity when the assumption of continuity breaks down and scales involved become of the order of the size of constituents. It is known from the literature on the rheology of the yield stress materials that non-local effects are present at such small scales. Non-local effects
impact significantly the flow properties and they can be reabsorbed into a continuum formulation by introducing an effective diffusivity in the dynamical equations for the “fluidity” field (i.e. inverse viscosity). If and how this is possible for the convective systems studied in this paper, certainly deserves future scrutiny.

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* francesca.pelusi@roma2.infn.it

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