

Competing Active and Passive Interactions Drive Amoeba-like Crystallites and Ordered Bands in Active Colloids

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Swimmers and self-propelled particles are physical models for the collective behaviour and motility of a wide variety of living systems, such as bacteria colonies, bird flocks and fish schools. Such artificial active materials are amenable to physical models which reveal the microscopic mechanisms underlying the collective behaviour. Here we study colloids in a DC electric field. Our quasi-two-dimensional system of electrically-driven particles exhibits a rich and exotic phase behaviour exhibiting passive crystallites, motile crystallites, an active gas, and banding. Amongst these are two mesophases, reminiscent of systems with competing interactions. At low field strengths activity *suppresses* demixing leading to motile crystallites. Meanwhile at high field strengths, activity *drives* partial demixing to travelling bands. We parameterise a particulate simulation model which reproduces the experimentally observed phases.

I. INTRODUCTION

From living organisms to synthetic colloidal particles, active systems display exotic phenomena not attainable by matter at thermal equilibrium [1–12], such as swarming [13, 14], cluster-formation [15–18], phase separation in the absence of attractions [6, 19–22], banding [23], and unusual crystallisation behaviour [24]. This is due to continuous energy consumption which occurs in a wide range of systems which can result in collective behaviour at very different lengthscales, from the cell cytoskeleton [25, 26], tissues [27] and bacterial colonies [28–31] to larger scales such as insect swarms [32], fish schools [33] and bird flocks [34]. Artificial active materials, composed of microswimmers, active colloids or vibrating granular particles [7, 15, 24, 29, 35], or even synthetically modified living systems such as bacteria [21, 36], provide a suitable testing ground where the behaviour of active matter may be carefully probed to extract the new physical principles of this class of matter.

While some progress has been made in the context of mapping to equilibrium behaviour [6, 19, 37–40], with notable exceptions [6, 41–43], theoretical approaches remain less developed. Key to the development of a theoretical understanding is to use simple models of active particles. While these capture some of the complex behaviour observed experimentally, for example collective motion and demixing [6, 41, 44–52], the link between experiment and theory in active matter is often rather qualitative. As a result, a comprehensive understanding of how and which microscopic mechanisms lead to the emergence of complex structures in experimental active systems remains elusive. Here we use particle-resolved studies to observe the Quincke roller system, active colloids which exhibit swarming and flocking. We parameterise our experimen-

tal system at the microscopic level of the interacting particles [53, 54]. While intriguing boundary phenomena are observed in this system [55], here we focus on bulk phase behaviour.

At low-to-moderate motility, we reveal the importance of competing passive interactions (long-ranged attractions) driving crystallisation and activity which leads to melting-like and evaporation-like behaviour. At high motility, the role of passive and active interactions is *reversed*: activity drives demixing resulting in a banding phase, whose ordered local structures result from the repulsive core of the particles. This competition between passive and active interactions is reminiscent of well-known passive systems with competing interactions such as amphiphiles, block copolymers and mixtures of charged colloids and non-absorbing polymer where competing interactions lead to modulated phases such as clusters and lamellae [56–59], which indeed resemble some structures we find here and which have been shown to persist in active systems [17, 41]. Our approach shows how one may use bottom-up designs of particulate active matter with precisely controllable macroscopic behaviour.

In the system we study, the application of a uniform DC electric field above a critical field strength E_Q induces *Quincke Rotation* of colloidal particles, which leads to directed motion by coupling their translation and diffusion near a surface [53, 60]. In the absence of a field, the particles behave as conventional passive Brownian colloids. At low field strengths, while remaining non-motile, particles agglomerate into crystals due to long-ranged attractive interactions which arise from electro-osmotic flows (Fig. 1A) [61–63]. Above E_Q , the particles undergo Quincke rotation [64–66] and become motile (Fig. 1B) so that the electro-osmotically generated crystallites transition into a highly mobile active state reminiscent of

amoebae (see Supplementary Movies 1-3 [86]).

Unlike “living crystals” [16], and systems exhibiting motility-induced phase separation [6, 18, 20, 21], here the aggregation is driven by long-ranged electrohydrodynamic interactions [61–63]. These “amoebae” are motile and characterised by a highly dynamic outer surface, and dissolve into an isotropic active gas as we increase the field strength. Finally at very high field strengths we find that the system transitions to an inhomogeneous polar state (“banding”) that has previously been investigated numerically and analytically in active matter systems [23, 44, 67–69], and experimentally observed [53, 70], but here we find a significant degree of local ordering. We investigate the rich structural and dynamical properties of our system using a range of static and dynamic order parameters. Central to our approach is to capture, quantitatively, the behaviour of the experimental system with a simulation model which captures the essentials of the system [53]. In this way, we provide the means to predict the behaviour of active matter systems in a rather accurate fashion.

II. METHODOLOGY

A. Experiments

A more extensive description of the experimental setup shown schematically in Fig. 1B. is included in the Appendix A. We use the so-called Quincke electrorotation mechanism of colloidal rollers [53]. A uniform electric field E is applied to the suspension. Above a critical field strength E_Q , the symmetry of the electric charge distribution at the colloid surface breaks spontaneously. As a result, an electric torque acting on the colloids leads to rotation with a constant rate around a random axis transverse to the field E (Fig. 1A) [64, 65]. Upon sedimentation, a quasi 2D system forms, and rotation couples with translation. As a result, roller motion along a random direction is observed.

We use suspensions of colloidal particles of diameter $\sigma = 2.92 \mu\text{m}$ in a non-aqueous ionic solution. Experiments are performed using sample cells made of two indium-tin-oxide (ITO) coated glass slides separated by double-sided tape. The ITO layers are used for the application of the electric field in the z -direction. Simultaneously, rollers are confined within a square region with the application of the field. This region is created by the removal of a photoresist layer, as illustrated in Fig. 1B. Note that a non-zero current develops solely within the square region of the cell. With Quincke rotation acting on the system, the roller trajectories are restrained to the confinement region. We translate the resultant field-dependent motion to dimensionless Péclet numbers Pe , which characterise the strength of the active motion with respect to the thermal diffusive (colloidal) motion. We henceforth characterise the static and dynamic behaviour of the system with increasing area fractions ϕ and field

strengths. Throughout, we use the Brownian time for a colloid to diffuse its own radius in 2D, $\tau = \sigma^2/D_t \approx 9$ s, as the unit of time, where D_t is the translational diffusion constant.

B. Simulations

The Quincke rollers are subject to forces and torques due to excluded volume repulsions, as well as self-propulsion, alignment and attractions generated by the electrohydrodynamic interactions of the particles with their environment and each other [53, 71]. They can be modelled as *active Brownian particles* with an additional *active aligning torque*, whose active/passive forces and torques can be quantitatively specified. We implement Brownian dynamics simulations, with the following equations of motion for positions and orientations \mathbf{r}_i, θ_i .

$$\dot{\mathbf{r}}_i = \frac{D_t}{k_B T} [\mathbf{F}_i + f^p \hat{\mathbf{P}}_i] + \sqrt{2D_t} \boldsymbol{\xi}_i^t, \quad (1)$$

$$\dot{\theta}_i = \frac{D_r}{k_B T} \mathcal{T}_i + \sqrt{2D_r} \xi_i^r, \quad (2)$$

where \mathbf{F}_i is the interparticle force on the i th roller, f^p is the magnitude of the active force, $\hat{\mathbf{P}}_i = (\cos \theta_i, \sin \theta_i)$ is the direction of motion of the i th roller, \mathcal{T}_i is the torque on the i th roller which incorporates alignment terms, and $\xi_i^{t,r}$ is a Gaussian white noise of zero mean and unit variance. D_r is the rotational diffusion constant. The direct interactions \mathbf{F}_i include a “hard” core and long-range attraction, the latter to model the electrohydrodynamic contribution. This pertains to long-ranged hydrodynamic attractions induced by solvent flow [61–63]. Further details of the model (which is partly based on Ref. [53]) and the simulation parameters, and the procedure by which the parameters were mapped to the experiment may be found in the Appendix. We emphasise that we use *one* set of interaction parameters to describe the simulations, that is to say, we only vary the area fraction and Péclet for all the state points studied.

C. Determining the Péclet number

Before moving to the discussion of our results, we first describe our mapping of field strength to Péclet number between experiment and simulation. We obtain the bare translational diffusion coefficient of the passive system D_t measured at equilibrium. Particle velocity v , and the characteristic time scale for the rotational diffusion $\tau_r = D_r^{-1}$ for a dilute sample with with area fraction $\phi \approx 0.001$ are obtained from the fitting to the mean square displacement (MSD) of active particles in the dilute (gas) regime,

$$\langle \Delta r^2(t) \rangle = 4D_t t + \frac{v^2 \tau_r^2}{3} \left[\frac{2t}{\tau_r} + \exp\left(\frac{-2t}{\tau_r}\right) - 1 \right]. \quad (3)$$

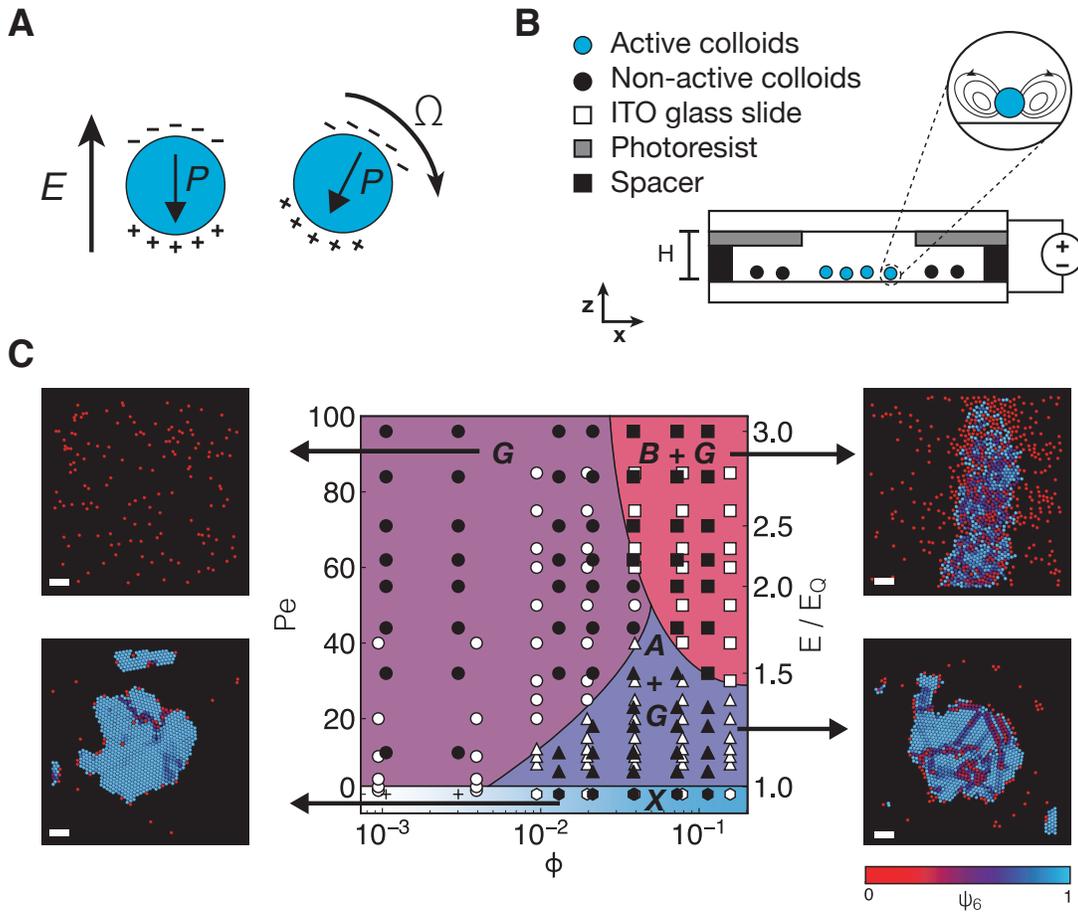


FIG. 1: Phase diagram of Quincke Rollers as a function of activity. **A. Schematic representation of mechanism of Quincke rolling.** The charge distribution around the sphere forms a dipole oriented inversely to the field direction, and any fluctuation in the dipole orientation leads to particle rotation with a constant angular speed Ω . The field-dependent activity is translated to the Péclet number, as described in the text. **B. Experimental set-up.** Colloidal particles are suspended within a sample cell made of conductive glass slides. Colloids are confined by an electrokinetic flow to the region of interest and they become active (blue particles). The induced electrohydrodynamic flow is represented by the solid lines in the amplified illustration. This flow field leads to long-ranged electrohydrodynamic interactions between the particles [71]. **C. Phase behaviour in the area fraction – Péclet number plane.** In the low activity regime, electrohydrodynamic interactions due to flow fields (see inset in Fig. 1B) result in (passive) crystallite formation at sufficient area fraction. On increasing the activity, e.g. $Pe = 2$ ($E = E_Q$, with $E_Q \approx 8 \times 10^5 \text{ V} \cdot \text{m}^{-1}$), the particles self-propel sufficiently that the dynamics change markedly (see Fig. S2B and Supplementary Movie 1 [86]), and the active crystals split and coalesce with one another. With a further increase of activity, the crystallites melt and we find polar bands propagating through active gas. Black and white symbols represent experimental and numerical data respectively. Symbols: \circ represent passive crystals (X), \circ active gas (G), \triangle active crystals and gas (A+G), \square bands and gas (B+G). Solid lines are drawn guides. Experimental snapshots for every phase in the diagram are indicated by arrows. Particles are coloured according to their hexagonal order parameter ψ_6 , whose magnitude is indicated by the colourbar. Scale bars represent $10 \mu\text{m}$.

To extract the parameters of Eq. 3 from the experiments we consider a series of similarly dilute samples at different field strengths. We estimate the dimensionless Péclet number as $Pe = 3v\tau_r/\sigma$, for each measured velocity in the different states obtained in the experiment. The Péclet number is defined in terms of Quincke rotation. However, since this is related to the threshold field strength E_Q where Quincke rotation is initiated, we find that for low field strengths, Pe is small and only weakly

dependent on the field, $E \ll E_Q$, $Pe \sim 0$. Once the particles become motile, for our system the two appear to be coupled for $E > E_Q$, as Pe scales with $[(E/E_Q)^2 - 1]^{1/2}$, (see Fig. S1 in the Supplemental Material [86]). Note that because the particles are colloidal, they can only be quasi 2D, and we expect slip boundary conditions between the particles and the substrate.

III. RESULTS

We now present our main findings. First we consider the phase behaviour of the system as a function of the activity, represented by the Péclet number which we obtain from measuring particle mobility, and as a function of area fraction. At zero field strength ($Pe = 0$), we obtain Brownian hard discs which form a 2D colloidal fluid for the area fractions we consider. Upon increasing the field strength for $\phi \gtrsim 0.03$, the system exhibits a novel phase behaviour owing to a coupling between non-equilibrium electrohydrodynamic interactions due to solvent flow and electrically induced activity (Quincke rotation). Then, by the use of a variety of dynamic and static order parameters, we identify the nature of the transitions between these states.

A. Crystallisation

At sufficient area fraction, we find that particle condensation to form crystallites emerges at low field strength, e.g. $Pe \approx 0$ ($E < E_Q$). This is due to the long-ranged electrohydrodynamic interactions (Fig. 1B) [71]. In our experiments, colloids act as dielectric regions perturbing the electric charge distribution, therefore inducing a flow of ions with a component tangential to the substrate [71]. In the vicinity of such an electro-osmotic flow, the particles experience transverse motion leading to the formation of crystallites (Fig. 1B). We find crystallisation for area fractions $\phi \gtrsim 10^{-2}$. We emphasise that this may be due to the finite size of our experimental cells. That is to say, even the passive system is likely out of equilibrium and for sufficient waiting time, we expect crystallite formation for $\phi \lesssim 10^{-2}$.

B. Activity-induced phase transitions

Upon increasing the field strength, we can exploit the Quincke mechanism that triggers spontaneous rotation (Fig. 1A) to study the behaviour of self-propelled rollers. For this to occur, the viscous torque acting on the particle must be overcome, hence the field needs to be sufficient to initiate rolling ($E \geq E_Q$). When increasing the activity above $Pe \approx 2$ ($E = E_Q$), we observe crystallite motility, that is to say, the crystallites are mobile by themselves, see Supplementary Movie 1, related to clustering behavior in Quincke rollers modified to exhibit run-and-tumble behaviour [72]. These active crystallites arise from the interplay of electrohydrodynamic interactions [61, 71] and the Quincke electro-rotation of the rollers. Note that for our system, the aggregation resulting from passive interactions does not fully suppress motility, as recently suggested for high-density active solids [55].

We further find coalescence and splitting of the crystallites, yet the local hexagonal symmetry remains, as can be seen in certain bacteria colonies [31] and chiral

swimmers [73]. We term this an “amoeba phase”, since the motility leads the aggregate to constantly reshape in a fashion reminiscent of the motion of amoebae, as shown by the time sequence in Fig. S2B (also see Supplementary Movie 1 [86]). These amoebae appear to be in a non-equilibrium steady state. However such an inhomogeneous state is reminiscent of mesophases in passive systems, but here activity suppresses demixing, playing the role of long-ranged repulsion in so-called “mermaid” systems [57–59, 74] and consistent with some recent predictions of active liquids [41]. We note that, for this emergence of motility, E is substantially larger than E_Q . There may be some suppression of the transition to Quincke rotation by the dense packing in the crystallites, but we caution that E_Q is approximate in any case.

On further increasing the field to $E = 1.75E_Q$ ($Pe = 44$), with $\phi \leq 4 \times 10^{-2}$ Quincke rotation triggers breakdown of the active crystallites into an ‘active gas’ of colloidal rollers undergoing displacement in random directions, Fig. 1C. Previously, it was shown experimentally that the increase in area fraction results in homogeneous polar phases and vortices [53, 54]. Here, we note that the onset to polar collective motion occurs experimentally with area fraction $\phi = 4 \times 10^{-2}$ and $Pe \geq 63$. Further increase in area fraction results in traveling bands through the gas at lower activity values, i.e. $Pe \approx 32$.

These bands form perpendicular to the direction of particle motion (which self-organizes into a strongly preferred direction) (see Supplementary Movies 4 and 5 [86]). This is related to banding observed in earlier experiments with Quincke rollers [53], but here the area fraction in the band is very much higher, leading to local hexagonal order (see Fig. 1C).

The traveling bands are akin to the liquid fractions in flocking models [69, 75], but here the high local area fraction leads to hexagonal order in the bands. However, within the activity and density values measured no homogeneous phase develops as in earlier experiments with Quincke rollers [53]. In our simulations, we see one band in the box. We leave the analysis of whether this is activity-driven micro-phase separation, or full demixing for a later finite-size scaling analysis. This local hexagonal order within the bands contrasts with the unstructured bands seen in the Vicsek model [23]. Here, it is the activity which *drives* the banding, i.e. particle demixing. This is quite unlike the case of the amoebae above, where activity *suppresses* full demixing.

C. Local Structure

Having qualitatively introduced the behaviour we encounter in our system in Fig. 1, we now proceed to consider the phase transitions in more detail. In order to determine the nature of the transitions we require suitable order parameters. We first consider the structural properties of the phases we encounter — passive fluid, passive crystal, active crystallite (“amoebae”), ac-

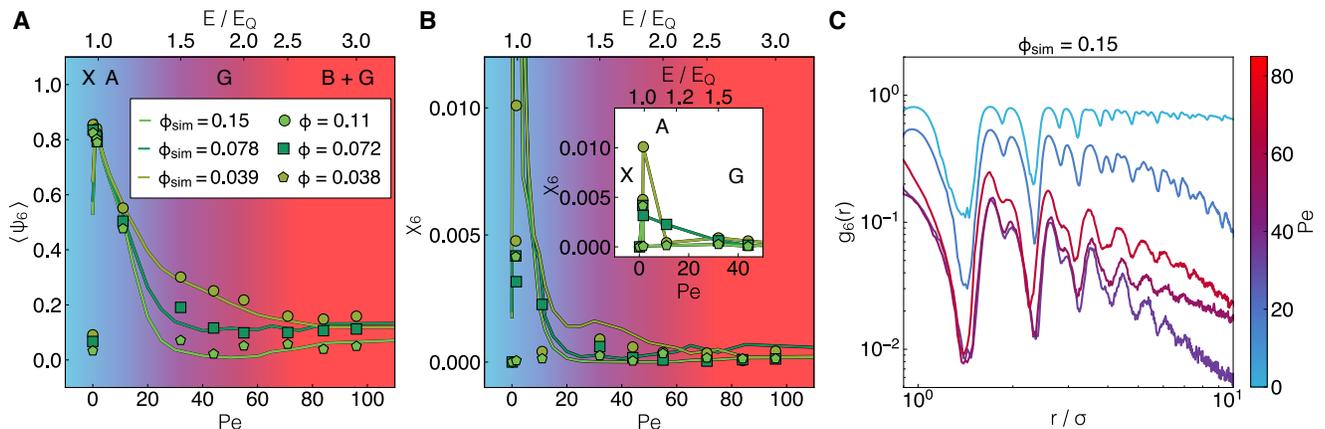


FIG. 2: **Changes in local structure as a function of field strength for Quincke rollers.** **A.** Local order determined with the bond-orientational parameter ψ_6 upon increasing Pe values. **B.** Fluctuations of the bond-orientational parameter χ_6 as defined in the text. Inset displays experimental measurements where a peak develops at the transition between the crystallites and the amoeba phase. Symbols and lines in **A** represent experimental and numerical data respectively for both **A** and **B**. **C.** Orientational correlation functions $g_6(r)$ for Pe as indicated in the color bar. Data obtained from simulations with $\phi = 0.15$. In **A** and **B** and in subsequent figures, the phases are denoted as X - crystallites, A - amoebae, B + G - (active) gas and banding.

active gas and bands. Given the richness of the phase behaviour, it is unlikely that one single order parameter will prove sufficient, and we find this to be the case. We begin with the 2D bond-orientational order parameter, $\psi_6 = (1/N) \sum_{i=1}^N |\psi_6^i|$. Perfect hexagonal ordering is indicated by $\psi_6 = 1$, whereas a completely disordered configuration gives $\psi_6 = 0$. See Appendix C for more details of ψ_6 .

In Fig. 2A, we plot the average ψ_6 as a function of Pe and the applied electric field for both experiment and simulation. We emphasise that, given the simplicity of our model, and of our mapping, the agreement between experiment and simulation is remarkable. We find low ordering of the passive Brownian system (at $E = 0$ or $Pe = 0$). With a slight increase in the field strength to $E < E_Q$, we observe a rapid rise in ψ_6 to ≈ 0.9 that corresponds to the crystallisation transition driven by the electrohydrodynamic interactions. In this regime, the system is composed of many crystallites that barely move. It is possible that there may be a condensed liquid (or hexatic) phase [76], although this is not apparent in our data, and the transition appears first-order within the field strengths we have sampled. We believe this to be similar to equilibrium 2D attractive systems undergoing crystallisation and move on to consider the activity-driven transitions.

Increasing the activity further into the amoeba phase, ψ_6 starts to decrease. However, ψ_6 remains significantly above zero indicating the amoeba clusters are crystal-like. While this state is far-from-equilibrium, the ψ_6 value exhibits temporal fluctuations consistent with a steady state (Fig. S2A [86]) and local order parameter ψ_6 reveals rotational motion of the amoebae. We infer that to distinguish the (passive) crystallites from the

amoebae, some kind of dynamic order parameter may prove suitable, and return to this below. At larger Pe ($11 \lesssim Pe \lesssim 40$), the value of ψ_6 , drops markedly, as the amoebae “dissolve”, apparently in a continuous fashion. Finally at very high Pe ($Pe \gtrsim 40$), we see the emergence of banding, a form of phase separation driven by activity. The value of ψ_6 again shows signs of increase for $\phi < 0.16$.

To gain further insight into these transitions, in Fig. 2B we plot the fluctuations in the hexatic bond-orientation order parameter which we take as $\chi_6 = \langle \psi_6^2 \rangle - \langle \psi_6 \rangle^2$ where the average is over different snapshots. Further details are provided in the Appendix C. At low Péclet numbers, we see good agreement between the experiment and simulation, but when the motility is higher, the simulations decay towards the active gas faster than the experiments. However, we find no enhancement in χ_6 around the amoeba-gas phase boundaries, indicating that the transition is a cross-over rather than a first-order-like transition between different phases.

To quantify the spatial correlations in ψ_6 , in Fig. 2C, we plot $g_6(r)$ defined as,

$$g_6(r = |\mathbf{r}_i - \mathbf{r}_j|) = \langle \psi_6^i \psi_6^j \rangle \quad (4)$$

where ψ_6^i is the (complex) value of the hexatic bond-orientation order parameter for particle i at position \mathbf{r}_i . At low Pe , we observe long-ranged orientational correlations in the crystal and amoeba regimes. Such correlations are significantly shorter-ranged for the active gas. Interestingly, for the largest Pe in the banding regime, we find that the bond-orientational order parameter is correlated over a larger domain than in the gas regime. Therefore, formation of the bands not only increases ψ_6 , but also enhances its spatial correlations.

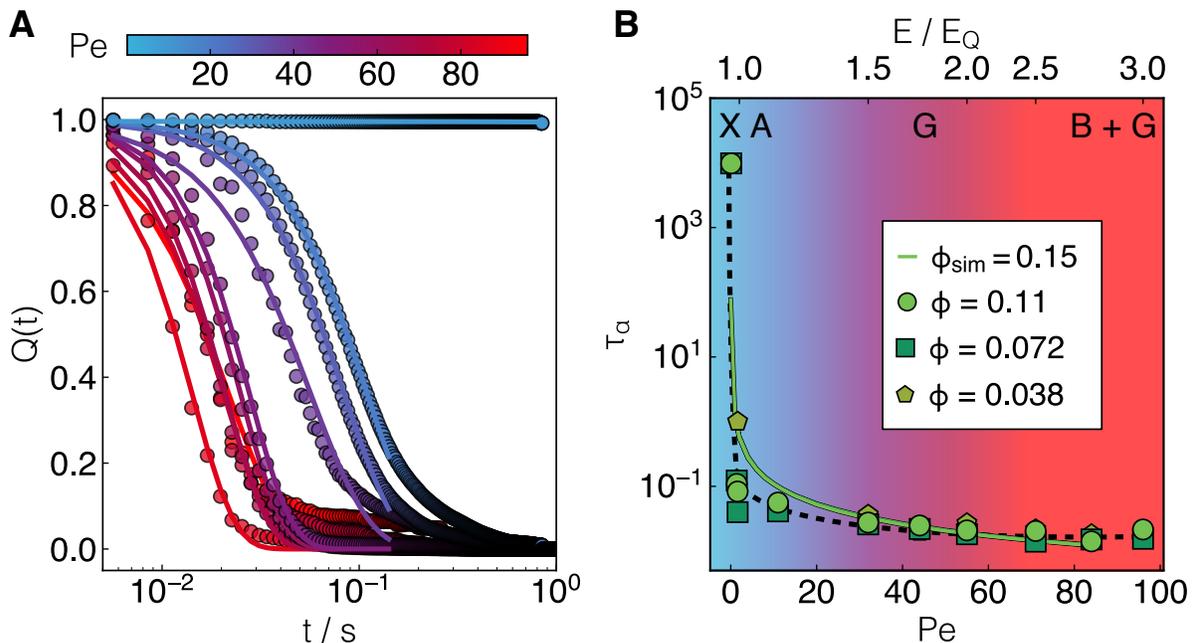


FIG. 3: **Dynamics of the Quincke Rollers across various phases.** **A.** Dynamical overlap function $Q(t)$, using Eq. 5. Symbols represent experimental data for $\phi = 0.11$. Solid lines are exponential fits as described in the text, where the stretching exponent b is constrained to 1 for both experiment and simulation. Color bar indicates the correspondent Pe for each line. Data are scaled by the Brownian time τ . **B.** Relaxation time τ_α from stretched-exponential fitting to symbols in **A**. Symbols represent experiments, solid line is obtained from simulations, and dashed line is a guide. The phases are denoted as X - crystallites, A - amoebae, B + G - (active) gas and banding.

D. Dynamical Analysis

In our analysis of the local structure in Fig. 2, we noted that some kind of dynamical order parameter would be appropriate to distinguish the crystallites from the amoebae. In Fig. 3, we use such an order parameter to perform this analysis, the overlap [24],

$$Q(t) = \left\langle \frac{1}{N} \sum_{i=1}^N \exp - \left(\frac{[\mathbf{r}_i(t'+t) - \mathbf{r}_i(t')]^2}{a^2} \right) \right\rangle_{t'} \quad (5)$$

which we evaluate at $a = \sigma$. We fit the resulting dynamic correlation functions with a stretched exponential form, where b is the stretching exponent, $Q(t) = \exp[-(t/\tau_\alpha)^b]$, as shown in Fig. 3A to determine a timescale for relaxation in our system, τ_α . We plot this timescale against the Péclet number in Fig. 3B.

Most striking in the crystal-amoeba transition is the massive drop in relaxation time, Fig. 3B: at a total of *five* decades, this is a very substantial dynamical change for particle-resolved studies of colloids, active or passive [77]. Thus the crystallites are effectively solids, while the amoebae exhibit timescales of colloidal liquids, even though their local structure is crystalline. Despite this precipitous drop in relaxation time, we find that the transition from crystallites to amoeba is apparently continuous in nature. We thus conclude that the

crystallite-amoebae and amoeba-active gas transitions we have found are both continuous, at least insofar as we can detect.

E. Characteristics of the Active and Passive Crystallites

In Fig. 2, the ψ_6 bond-orientational order parameter gave somewhat limited insight as to the nature of the crystallite-amoeba transition, as both exhibit hexagonal local symmetry. Therefore we now seek other structural measures. Figure 4A shows how the mean cluster size varies in different regimes. We consider four particles as the minimum cluster size. The system is composed of a few large clusters at very low Pe . Upon increasing the activity, those big clusters break up to smaller ones until in the inhomogeneous regime where the system is dominated by monomers and small clusters from collisions. Hence, the low mean size in Fig. 4A, despite the emergence of denser bands. Note that in the regime where our simulations indicate banding, finite size effects in the simulations (which have 10000 particles) may influence the cluster size somewhat as the bands span the simulation box. The same holds for the passive crystals at low field strength.

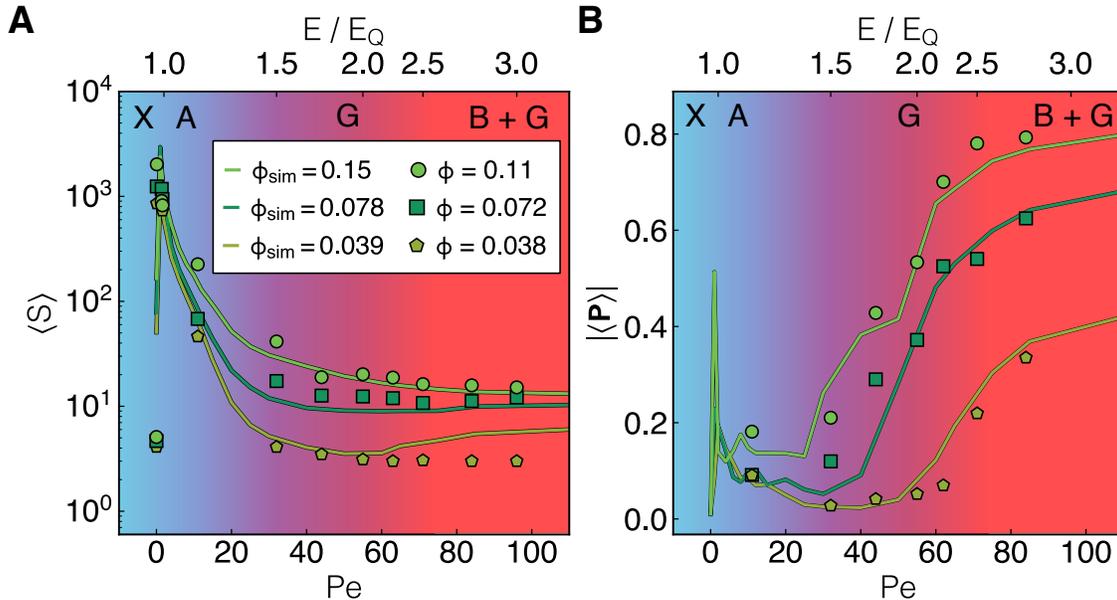


FIG. 4: **Characteristics of the clusters formed by the Quincke rollers.** **A.** Size of clusters as a function of activity. Rapid increase is observed as crystallites form. Symbols represent experiments and lines are from numerical analysis. **B.** Polar order for different Pe values. Symbols are data obtained from many velocity measurements over different regions in the sampling cell. As above, the phases are denoted as X - crystallites, A - amoebae, B + G - (active) gas and banding.

F. Nature of the Transitions at Higher Activity: Amoeba to Active Gas and Active Gas to Ordered Bands

In addition to the transitions we have already discussed, we encounter more at higher field strength. Firstly, the amoebae “dissolve” to form an “active gas”. At the densities we consider, this transition is characterised by a substantial – but continuous – drop in the ψ_6 bond-orientational order parameter (Fig. 2A) consistent with our discussion of the continuous change in dynamics above.

At higher field strengths, we encounter banding, strong density fluctuations perpendicular to the preferred direction of travel. Interestingly, this inhomogeneous state exhibits some degree of local order, as the value of the bond-orientational order parameter $\langle \psi_6 \rangle \approx 0.2$. At higher densities, e.g. $\phi \gtrsim 10^{-1}$, the local order of dense bands is notably higher, with $\langle \psi_6 \rangle > 0.5$ (see B+G panel Fig. 1C). While far from indicating full hexagonal order ($\langle \psi_6 \rangle = 1$), this is nevertheless significantly larger than zero. Furthermore, as we can see in Fig. 1C, some particles are in a very high state of crystalline order (appearing blue), although most are not. Previous work did not observe hexagonal ordering [53, 54], we believe this is due to the fact the bands in our case form at much higher area fraction, such that excluded volume effects contribute to the ordering. We note that here we use polymethylmethacrylate particles, while Bricard *et al.* [53, 54] used polystyrene particles whose electrostatic charging properties may be different.

Rather striking, in the case of the transition to the banded phase is the alignment between the dipoles of the Quincke rollers, which defines the direction of rotation. In Fig. 4B, we see a very strong increase in the alignment upon banding, suggesting that this is a suitable order parameter in this case. Taking the polarisation $|\langle \mathbf{P} \rangle|$ as an order parameter for the transition between active gas and bands, we find that it is continuous.

IV. DISCUSSION

We have shown that the Quincke roller system exhibits a rich and complex phase behaviour, with passive fluid, crystal, amoeba-like active crystallites, active gas and an ordered banding phase. We reveal an intriguing reversal in the roles of active and passive interactions. At low field strength, activity suppresses demixing, while (passive) electrohydrodynamic interactions drive partial demixing in the “amoeba” phase. At high field strength, the situation is quite reversed: here activity drives partial demixing into bands. This is consistent with recent theoretical predictions for active liquids [41] and the well-known phenomenon of motility-induced phase separation [6].

We have used a variety of static and dynamic order parameters to probe the nature of the transitions between these states, and find that they are continuous in nature except the (passive) fluid-crystal transition which is consistent with first-order. One intriguing question concerns the universality of such states in active matter with at-

tractions, and the role of hydrodynamics [78].

For the simulation model, we have quantitatively parameterised the components of the Quincke roller system by treating the electrohydrodynamic attraction with a long-ranged potential, “hard” core, active force and electrohydrodynamic alignment terms. Remarkably, when we rescale our results to compare the same Péclet numbers in experiments and simulations, we obtain a quantitative agreement between the two. With the model, we have revealed that a key ingredient of the phase behaviour is the interplay between *active* and *passive* interactions.

To date, there are few examples of quantitative agreement between particular models and experiments in active matter, and it is in this quantitative agreement between simulation and experiment that our work is significant. Our work opens the way to using simple, intuitive minimal models which correctly capture the *microscopic* interactions to describe, quantitatively, the *macroscopic* physical behaviour of complex active systems which are far-from-equilibrium.

Appendix: Materials and Methods

A. Experimental setup

Our experimental Quincke roller model consists of poly(methyl methacrylate) (PMMA) spheres of diameter, $\sigma = 2.92 \mu\text{m}$ determined with SEM. These are suspended in a 0.15 mM solution of dioctyl sulfosuccinate sodium (AOT) in hexadecane. Imaging and application of a uniform DC field take place in sample cells made of two indium tin oxide (ITO)-coated glass slides (Solems ITOSOL12), separated with a layer of adhesive tape of thickness $H = 100 \mu\text{m}$. Additionally, a layer of photoreist (Microposit S1818) of $2 \mu\text{m}$ in thickness is deposited on the top electrode. Square confinement regions of $5 \text{ mm} \times 5 \text{ mm}$ are created using conventional lithography techniques. The same electric field E that triggers Quincke rotation induces a lateral electric potential gradient between the conductive region and the insulating photoreist layer. As a result, an electrokinetic inward flow confines the rollers at the bottom electrode [61]. The electric field is applied by a power supply (Elektro Automatik, PS-2384-05B) and amplified (Trek 606E-6). Image sequences are obtained using brightfield microscopy (Leica DMI 3000B) with a 10x objective and recorded with a frame rate of 354 fps (Basler Ace). All measurements were performed when the rollers reached a steady state. Individual rollers are identified and particle trajectories are reconstructed using a Python version of conventional tracking methods [79].

B. Determination of the critical strength

We follow the description of Lemaire and co-workers [65, 80] to estimate the critical field strength, E_Q . The spontaneous rotation of particles, known as Quincke rotation, strongly depends on the charge distribution at the particle-liquid interface and the respective charge relaxation times, given by $\tau_{p,l} = \epsilon_{p,l}/s_{p,l}$, where $\epsilon_{p,l}$ and $s_{p,l}$ are the dielectric constant and conductivity of the particle and the liquid respectively. In the case of having $\tau_l > \tau_p$, the induced dipole \mathbf{P} is stable with respect of field direction. On the other hand with $\tau_p > \tau_l$, \mathbf{P} is unstable with respect to the field direction (see Fig. 1A in main text), and any perturbation results in an electrostatic torque $\mathcal{T}^e = \mathbf{P} \times \mathbf{E}$, from the dipole rotation. Nevertheless, even if $\tau_p > \tau_l$ is satisfied, \mathcal{T}^e needs to overcome the viscous torque exerted on the particle by the liquid to initiate rotation, $\mathcal{T}^H = -\alpha\omega$, where the angular velocity is given by ω and $\alpha = \pi\eta\sigma^3$ is the rotational friction coefficient. We use polymethyl methacrylate colloids of diameter $\sigma = 2.92 \mu\text{m}$, with $\epsilon_p = 2.6\epsilon_0$, and a 0.15 mM AOT/hexadecane solution with $\eta = 4.3 \text{ mPa}\cdot\text{s}$, $s_l \approx 10^{-8} \text{ S m}^{-1}$ [81], $s_p \approx 10^{-14} \text{ S m}^{-1}$ [65] and $\epsilon_l \approx 2\epsilon_0$ for our system. The critical threshold is given by

$$E_Q = \left[\frac{1}{2} \pi \epsilon_l \sigma^3 (\mathcal{P}^0 - \mathcal{P}^\infty) \tau_{\text{MW}} \alpha^{-1} \right]^{-1/2}, \quad (6)$$

where the polarisability factors

$$\mathcal{P}^0 = \frac{s_p - s_l}{s_p + 2s_l} \quad (7)$$

and

$$\mathcal{P}^\infty = \frac{\epsilon_p - \epsilon_l}{\epsilon_p + 2\epsilon_l} \quad (8)$$

account for the conductivities and permittivities of the particle and liquid respectively. The characteristic dipole relaxation timescale is given by the Maxwell-Wagner time,

$$\tau_{\text{MW}} = \frac{\epsilon_p + 2\epsilon_l}{s_p + 2s_l}. \quad (9)$$

C. Order parameter details

Here we take the mean of the bond-orientational order parameter ψ_6 across N particles

$$\psi_6 = \frac{1}{N} \sum_{j=1}^N |\psi_6^j|. \quad (10)$$

The value of the order parameter for each particle is

$$\psi_6^j \equiv \frac{1}{Z_j} \sum_{k=1}^{Z_j} \exp(i6\theta_k^j) \quad (11)$$

where Z_j is the co-ordination number of particle j obtained from a Voronoi construction and θ_k^j is the angle made between a reference axis and the bond between particle j and its k th neighbour. $\psi_6 = 1$ indicates perfect hexagonal ordering, whereas completely disordered structures give $\psi_6 = 0$. Fig. 2A shows that for a passive Brownian system there is almost no hexagonal order.

We quantify the fluctuations in ψ_6 by defining the susceptibility

$$\chi_6 \equiv \langle \psi_6^2 \rangle - \langle \psi_6 \rangle^2 \quad (12)$$

where $\psi_6^2 = 1/N \sum_{j=1}^N |\psi_6^j|^2$.

D. Simulation details

Brownian dynamics simulations were performed on a 2D system composed of $N = 10000$ interacting Quincke rollers. We integrate the over-damped Langevin equations (Eqs. 2) using the stochastic Euler scheme with a time step of $dt = 10^{-5}\tau$. In our simulations, the interparticle force on the i th roller $\mathbf{F}_i = -\nabla_i(\mathcal{H}_{\text{attr}} + \mathcal{H}_{\text{exc}})$ while the torque on the i th roller $\mathcal{T}_i = -\partial\mathcal{R}_{\text{align}}/\partial\theta_i$. The particle diameter σ , thermal energy $\epsilon = k_B T$ and Brownian time $\tau = \sigma^2/D_t$ are chosen as basic units for length, energy and time, respectively. We take $D_r = 3D_t/\sigma^2$, as expected for a spherical particle in the low-Reynolds-number regime. We study the phase behaviour of the system as a function of two dimensionless parameters; Péclet

number $Pe = f^p\sigma/k_B T$ and the area fraction $\phi = \frac{N\pi\sigma^2}{4L^2}$, where L is the linear size of the simulation box.

E. Microscopic Model of Alignment Interactions in Quincke Rollers

The following description is based on a microscopic model describing the dynamics of a population of colloidal rollers due to Quincke rotation. The direct interactions are captured in the force \mathbf{F}_i in Eq. 13. Here we consider the alignment terms. The equations of motion for the i th self-propelled particle are given by the following Langevin equations, where for the rotational case we have rewritten the version in the main text to explicitly consider the effective alignment interaction.

$$\dot{\mathbf{r}}_i = \frac{D_t}{k_B T} [\mathbf{F}_i + f^p \hat{\mathbf{P}}_i] + \sqrt{2D_t} \boldsymbol{\xi}_i^t \quad (13)$$

and

$$\dot{\theta}_i = -\frac{D_r}{k_B T} \frac{\partial}{\partial \theta_i} \sum_{j \neq i} \mathcal{R}_{\text{align}}(\mathbf{r}_{ij}, \hat{\mathbf{P}}_i, \hat{\mathbf{P}}_j) + \sqrt{2D_r} \xi_i^r \quad (14)$$

where the particle i is subject to a propulsion force of magnitude f^p whose direction changes due to the alignment interaction and noise ξ_i . We consider a pairwise alignment interaction between rollers that leads to a torque on particle i . Note that because the simulations are strictly in 2D, the direction of the dipole \mathbf{P} in Eq. 14 is that of the rotation, *i.e.* the direction of self-propulsion, rather than the (3D) induced dipole of the experimental system mentioned above.

Introduced by Caussin and Bartolo [53], the effective alignment interaction $\mathcal{R}_{\text{align}}$ reads

$$\mathcal{R}_{\text{align}}(\mathbf{r}, \hat{\mathbf{P}}_i, \hat{\mathbf{P}}_j) = -A_1(r) \hat{\mathbf{P}}_i \cdot \hat{\mathbf{P}}_j - A_2(r) \hat{\mathbf{r}} \cdot (\hat{\mathbf{P}}_i - \hat{\mathbf{P}}_j) - A_3(r) \hat{\mathbf{P}}_j \cdot (2\hat{\mathbf{r}}\hat{\mathbf{r}} - \mathbf{I}) \cdot \hat{\mathbf{P}}_i, \quad (15)$$

where $\hat{\mathbf{P}}_i = (\cos\theta_i, \sin\theta_i)$ is the direction of motion of the i th roller, and $\hat{\mathbf{r}} \equiv \mathbf{r}/r$. This has the minimum number of terms required to describe the electrohydrodynamically induced alignment interactions with the correct symmetry and whose range is set by the distance between plates in the experimental setup. We truncate

$\mathcal{R}_{\text{align}}$ at $r_{c1} = 3.0\sigma$, where σ is the particle diameter. We note that angular momentum is not conserved by these dynamics.

The coefficients $A_1(r)$, $A_2(r)$ and $A_3(r)$ incorporate the microscopic parameters, and are given by:

$$A_1(r) = 3\tilde{\mu}_s \frac{\sigma^3}{8r^3} \Theta(r) + 9 \left(\frac{\mu_{\perp}}{\mu_r} - 1 \right) \left(\mathcal{P}^{\infty} + \frac{1}{2} \right) \left(1 - \frac{E_Q^2}{E^2} \right) \frac{\sigma^5}{32r^5} \Theta(r) \quad (16. 1)$$

accounting for the short-ranged hydrodynamic inter-

actions and electrostatic couplings that promote the

alignment of directions between particles i and j . Here, μ_{\perp} and μ_r are the mobility coefficients depending on the liquid viscosity and the distance d between the surface and particle respectively. From the expressions in [82–85] we obtain $\mathcal{P}^{\infty} = 0.08$, $\tilde{\mu}_s = 11$ and $\mu_{\perp}/\mu_r = 1.5$.

The electrostatic repulsion and the electrohydrodynamic interactions coupling are encoded in the $A_2(r)$ and $A_3(r)$ coefficients respectively,

$$A_2(r) = 6 \left(\frac{\mu_{\perp}}{\mu_r} - 1 \right) \sqrt{\frac{E^2}{E_Q^2} - 1} \left[\left(\mathcal{P}^{\infty} + \frac{1}{2} \right) \frac{E^2}{E_Q^2} - \chi^{\infty} \right] \frac{\sigma^4}{16r^4} \Theta(r) \quad (16.2)$$

$$A_3(r) = 2\tilde{\mu}_s \frac{\sigma^2}{4r^2} \frac{\sigma}{2H} + \left[\tilde{\mu}_s \frac{\sigma^3}{8r^3} + 5 \left(\frac{\mu_{\perp}}{\mu_r} - 1 \right) \left(\mathcal{P}^{\infty} + \frac{1}{2} \right) \left(1 - \frac{E_Q^2}{E^2} \right) \frac{\sigma^5}{32r^5} \right] \Theta(r) \quad (16.3)$$

where the hydrodynamic and electrostatic couplings are screened over distances proportional to the chamber distance, $H = 100 \mu\text{m}$. A more detailed description can be found in Refs. [53], and [54]. We estimate such coefficients considering the experimental field intensity under which we observe the active gas phase ($E \geq E_Q$, with $E_Q \approx 8 \times 10^5 \text{ V} \cdot \text{m}^{-1}$), and average them over distances $r \in [\sigma, 3\sigma]$. For convenience we approximate the screening function as $\Theta(r) = 1$ if $r \leq H/\pi$ and $\Theta(r) = 0$ otherwise.

The interparticle force on the i th roller reads $\mathbf{F}_i = -\nabla_i(\mathcal{H}_{\text{attr}} + \mathcal{H}_{\text{exc}})$, where the electro-osmotic long-ranged attraction [71] is modelled by a truncated and shifted (at $r_{c2} = 5.0\sigma$) potential of the form

$$\mathcal{H}_{\text{attr}} = -A_4 \exp(-\kappa r)/r^2, \quad (17)$$

where $\kappa = 1/3\sigma^{-1}$ is the inverse screening length. The excluded volume interactions between rollers are represented by a repulsive Weeks-Chandler-Anderson (WCA) interaction of the form $\mathcal{H}_{\text{exc}} = 4\epsilon((\sigma/r)^{12} - (\sigma/r)^6) + \epsilon$, where $\epsilon = k_B T$ is the energy unit of the model. The WCA potential is truncated at $r_{c3} = 2^{1/6}\sigma$.

The coupling parameters in the alignment interactions are estimated to be $A_1 = 0.93k_B T$, $A_2 = 0.33k_B T$ and

$A_3 = 0.48k_B T$ for our experimental conditions, and we chose the attraction strength to be $A_4 = 10k_B T$. We verified that the qualitative phase behaviour of the model remains the same if we vary the strength of the long-ranged attraction. We note that we have parameterised A_1, A_3 from the single particle dynamics in the dilute gas phase, the attractive interactions A_2, A_4 are determined from the experimental parameters.

F. Supplementary Materials

Acknowledgements

The authors would like to thank Denis Bartolo, Olivier Dauchot, Jens Eggers, Mike Hagan, Rob Jack, Cristina Marchetti, Sriram Ramaswamy, Thomas Speck and Chantal Valeriani for helpful discussions. CPR, JH and FT would like to acknowledge the European Research Council under the FP7 / ERC Grant agreement n° 617266 “NANOPRS”. AMA is funded by CONA-CyT. TBL and MM are supported by BrisSynBio, a BBSRC/EPSRC Advanced Synthetic Biology Research Center (grant number BB/L01386X/1). Part of this work was carried out using the computational facilities of the Advanced Computing Research Centre, University of Bristol.

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