Active Model H: Scalar Active Matter in a Momentum-Conserving Fluid

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We present a continuum theory of self-propelled particles, without alignment interactions, in a momentum-conserving solvent. To address phase separation, we introduce a dimensionless scalar concentration field ϕ with advective-diffusive dynamics. Activity creates a contribution $\Sigma_{ij} = -\hat{\kappa}[(\partial_i\phi)(\partial_j\phi) - (\nabla\phi)^2\delta_{ij}/d]$ to the deviatoric stress, where $\hat{\kappa}$ is odd under time reversal and d is the number of spatial dimensions; this causes an effective interfacial tension contribution that is negative for contractile swimmers. We predict that domain growth then ceases at a length scale where diffusive coarsening is balanced by active stretching of interfaces, and confirm this numerically. Thus, there is a subtle interplay of activity and hydrodynamics, even without alignment interactions.

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"Active matter" means the collective dynamics of self-propelled particles. By converting energy into motion, such particles violate time-reversal symmetry (TRS) at the microscale, allowing far-from-equilibrium physics such as spontaneous flow at scales much larger than the particle size or spacing [1]. Active matter can be "wet," i.e., coupled in bulk to a momentum-conserving solvent, or "dry," i.e., in contact with a momentum-absorbing medium [2]. Wet active systems include bacterial swarms in a fluid, the cytoskeleton, biomimetic cell extracts [1,3–5], and synthetic colloidal swimmers in a fully bulk geometry [6]. The latter may, in the future, offer a toolbox for directed assembly of new materials.

Of particular importance is "active liquid crystal" (ALC) theory [1,7], which starts from a passive fluid of rodlike objects [8] with either a polar order parameter $\mathbf{P}$ [9], or a nematic one $\mathbf{Q}$ [10]. An active stress $-\zeta\mathbf{P}\otimes\mathbf{P}$ or $-\zeta\mathbf{Q}$ is then added, with ζ odd under time reversal and the dyadic product $\otimes$, representing the leading-order TRS violation in an orientationally ordered medium. This causes new physics such as giant number fluctuations [11] and spontaneous flow [9,10]. The latter instability depends on whether particles are extensile (pulling fluid inward equatorially and emitting it axially) or contractile (vice versa). Numerical solutions [12–14] resemble experiments on bacterial swarms [3] and microtubule-based cell extracts [5].

There is one important effect of activity that ALC models do not capture (unless added manually [15]): motility-induced phase separation (MIPS) [16,17]. If their propulsion speed $w(\rho)$ falls fast enough with the local density ρ (e.g., due to collisions), even purely repulsive active particles phase-separate into dense and dilute domains. MIPS is, by now, well-established, at least for dry models such as Brownian dynamics simulations of active spheres [18–21], and is an important benchmark problem for continuum theories. Unlike the physics of ALCs, MIPS does not depend on alignment interactions [18] and can be captured at continuum level by a dimensionless scalar concentration field ϕ alone, without direct appeal to $\mathbf{P}$ or $\mathbf{Q}$ [20,22]. This is pertinent to spherical colloidal swimmers, like many artificial microswimmers [6], which (although not entirely devoid of alignment interactions [23,24]) are, in contrast to ALCs, not orientationally ordered in the passive limit. The theory of "scalar active matter," with MIPS as its main feature, has been developed, so far, only for dry systems [16,17,25,26]. Following the same path as for passive systems [27,28] culminates in a dynamical scalar ϕ^4 field theory, called "Active Model B" [22].

In this Letter, we extend this simplest scalar active model to the wet case, complementing previous work on hydrodynamic interactions among individual spherical swimmers [23,24], and address phase separation as a benchmark problem. For passive systems, it is well known how to couple a diffusive, conserved, phase-separating order-parameter field $\phi(\mathbf{r}, t)$ (see passive Model B [27]) to an isothermal fluid flow; the result is called "Model H" [27]. We follow a similar path but find that activity deeply alters the relation between the stress and the scalar order parameter. At first sight, our "Active Model H" resembles closely passive Model H, in which the domain size $L(t) \propto \sigma t/\eta$ scales linearly in time, as found from a force balance between interfacial tension σ and dynamic viscosity η. On closer inspection though, our model involves two separate tensions, one in the diffusive and one in the mechanical sector. The second can be (and for pure MIPS is) negative for contractile particles. Therefore, while extensile systems show relatively normal domain growth, contractile ones should cease to coarsen at a certain scale, set by a balance between loss of interfacial area through diffusive coarsening and its creation through the contractile stress. The latter effect arises because microswimmers are more likely to

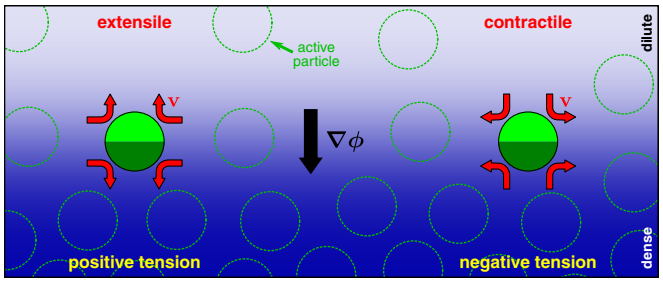

FIG. 1 (color online). Schematic showing the flow caused by swimmers (full spheres) whose polarization is normal to an interface between dense (dark background) and dilute (light background) phases. For contractile swimmers this is mechanically equivalent to a negative interfacial tension.

point normal to an interface than tangential to it (see Fig. 1). This orientational bias has a purely kinematic explanation: it does not require interparticle torques.

Active Model B.—For dry scalar active matter, the diffusive dynamics of $\phi(\mathbf{r},t)$ obeys [22,29]

$$\dot{\phi} = -\nabla\cdot\mathbf{J}, \tag{1}$$

$$\mathbf{J} = -\nabla\mu + \mathbf{\Lambda}, \tag{2}$$

$$\mu = a\phi + b\phi^3 - \kappa\nabla^2\phi + \lambda(\nabla\phi)^2. \tag{3}$$

Here, $\mathbf{J}$ is the diffusive current with unit mobility, $\mathbf{\Lambda}$ is a standard Gaussian white noise, and a, b, κ, and λ are constants ($a<0$ to ensure phase separation, $b>0$ and $\kappa>0$ for stability). This Active Model B differs from the traditional passive Model B [27] solely by addition of the leading-order TRS violation $\lambda(\nabla\phi)^2$. Without this term, μ, which resembles a chemical potential, can be written as $\delta\mathcal{F}/\delta\phi$ with the functional (setting $k_BT=1$)

$$\mathcal{F} = \int\left[\frac{a}{2}\phi^2 + \frac{b}{4}\phi^4 + \frac{\kappa}{2}(\nabla\phi)^2\right]\mathrm{d}^d r. \tag{4}$$

For active systems in d spatial dimensions, $\mathcal{F}$ is not a genuine free-energy functional, and in the simplest MIPS theory, it stems solely from the density-dependent speed $w(\rho)$ [16]. Nonetheless, for $\lambda=0$ Eqs. (1)–(3) coincide with Model B, which describes a passive system with free energy $\mathcal{F}$ [27]. In contrast, for $\lambda\neq 0$, no functional $\mathcal{F}$ exists for which $\mu=\delta\mathcal{F}/\delta\phi$ [22]. Thus, only the λ term allows a steady-state flux to arise, thereby violating TRS in the macroscopic equations, even if the physical origins of a, b, and κ also do so microscopically.

Active Model H.—Now, we couple the diffusive dynamics of $\phi(\mathbf{r},t)$ to a momentum-conserving solvent with fluid velocity $\mathbf{v}(\mathbf{r},t)$. Diffusive dynamics now takes place in the frame of the moving fluid so that Eq. (1) acquires an advective time derivative,

$$\dot{\phi} + \mathbf{v}\cdot\nabla\phi = -\nabla\cdot\mathbf{J}, \tag{5}$$

with no change to Eqs. (2) and (3). The fluid is incompressible,

$$\nabla\cdot\mathbf{v} = 0, \tag{6}$$

and of unit mass density. The Navier-Stokes equation for momentum conservation then reads

$$\dot{\mathbf{v}} + \mathbf{v}\cdot\nabla\mathbf{v} = \eta\nabla^2\mathbf{v} - \nabla p + \nabla\cdot\mathbf{\Sigma}, \tag{7}$$

where the pressure field $p(\mathbf{r},t)$ subsumes all isotropic stress contributions and enforces Eq. (6); the deviatoric stress $\mathbf{\Sigma}$ is traceless and (without orienting interactions) symmetric. In passive systems, $\mathbf{\Sigma}$ can be derived from $\mathcal{F}$ by standard procedures [31]; restoring isotropic terms, one finds $\nabla\cdot\mathbf{\Sigma} = -\phi\nabla\mu$, which is the thermodynamic force density arising from concentration gradients [28]. The deviatoric stress is then

$$\Sigma_{ij} = -\hat{\kappa}\left[(\partial_i\phi)(\partial_j\phi) - \frac{1}{d}(\nabla\phi)^2\delta_{ij}\right], \tag{8}$$

with $\hat{\kappa}=\kappa$. But, if we allow for general $\hat{\kappa}$, Eq. (8) remains the only deviatoric stress that can be created from $\phi(\mathbf{r},t)$ to order $\mathcal{O}(\phi^2,\nabla^3)$ [32], and is, hence, the sole leading-order deviatoric stress contribution for scalar active matter [33]. This neither contradicts, nor depends on, recent analyses of the pressure and/or stress in specific active models [35–39]. Our Active Model H comprises Eqs. (2), (3), and (5)–(8). Regardless of the underlying cause of phase separation (conventional attractions and/or MIPS in any combination), only the activity-dependent parameters λ and $\hat{\kappa}$ distinguish Active Model H from a passive Model H with the free-energy functional (4).

Our previous work on Active Model B shows λ to have benign effects on dynamics: its main effect is to shift the coexistence condition between phases [20–22]. Standard diffusive dynamics (e.g., Ostwald ripening [28]) holds qualitatively; although simulations show deviations from the expected power law $L\propto t^{1/3}$, these likely represent a slow crossover to that asymptote [22]. Below we focus mainly on the role of $\hat{\kappa}$.

This can be addressed in two contrasting limits. Suppose first we have a system with strong interparticle attractions, which would phase separate without activity. Then the thermodynamic, zeroth-order result is $\hat{\kappa}=\kappa$ as mentioned previously. Perturbative activity will change this only slightly. TRS is broken, but as with λ the effect on phase ordering dynamics should be benign. Specifically, we can use standard power-counting arguments [28,40] to estimate the dynamics of the domain size $L(t)$. Setting $\lambda=0$ for simplicity and ignoring inertia (which leads to a third regime at very late times [40,41]), we find [30]

$$\dot{L} \simeq \alpha\sigma/L^2 + \beta\tilde{\sigma}/\eta, \tag{9}$$

with dimensionless constants α and β. $\sigma=(-8\kappa a^3/9b^2)^{1/2}$ is the interfacial tension in ϕ^4 theory. When activity is nonzero but small, $\hat{\kappa}\neq\kappa$ and the tension $\tilde{\sigma}$ differs slightly from σ. The first contribution to $\dot{L}$ is diffusive (so $\dot{L}$ is a flux

$\propto \nabla\mu \sim \Pi/L \sim \sigma/L^2$ with the Laplace pressure Π, where σ is unperturbed since $\lambda = 0$); $\hat{\kappa}$ does not enter here. The second is hydrodynamic (balancing a viscous stress $\eta\nabla^2 v \sim \eta\dot{L}/L^2$ against $\nabla\cdot\mathbf{\Sigma} \sim \tilde{\sigma}/L^2$). When $\tilde{\sigma} = \sigma$, Eq. (9) captures the well-studied crossover from Model B behavior $L \propto (\sigma t)^{1/3}$ to viscous hydrodynamic (VH) coarsening $L \propto \tilde{\sigma} t/\eta$ at a length scale $L_\times \propto \eta^{1/2}$. Despite loss of TRS, a perturbative shift in $\tilde{\sigma}$ is unlikely to change this outcome, since Eq. (9) rests only on dimensional analysis [28].

The opposite limit is that of pure MIPS, where there are no attractions between particles and every coefficient in Active Model H is set by far-from-equilibrium physics [primarily, by $w(\rho)$ [16,20]]. In a TRS system, the form of $\mathcal{F}$ stems from a Hamiltonian that determines the diffusive (from $\nabla\mu$) and mechanical (from $\mathbf{\Sigma}$) currents in a thermodynamically consistent way. However, as already mentioned, in MIPS, $\mathcal{F}$ is merely a mathematical construct; there is no Hamiltonian (even for $\lambda = 0$) and, hence, no link between κ and $\hat{\kappa}$. Indeed, whereas κ is always positive, $\hat{\kappa}$ can have either sign as we now show.

The argument is based on simple kinematics. For a system of swimmers with a propulsion speed $w(\rho)$ that depends on local particle density $\rho(\mathbf{r}, t)$ but not on swimming direction $\hat{\mathbf{u}}$, the first and second orientational moments of the distribution function $\Psi(\mathbf{r}, \hat{\mathbf{u}}, t)$ obey [17]

$$\mathbf{P} = -\tau\nabla(w\rho), \tag{10}$$

$$\mathbf{Q} = -\tau\frac{d-1}{2d}\nabla(w\mathbf{P})^{\mathrm{ST}}. \tag{11}$$

Here, τ is the orientational relaxation time; ρ is the zeroth orientational moment of Ψ; and ST denotes the symmetric traceless component. Note that ρ and ϕ are related linearly; specifically, if one sets $a = -b = -1$, one has $\phi = (2\rho - \rho_H - \rho_L)/(\rho_H - \rho_L)$, where ρ_L and ρ_H denote the low and high densities of coexisting phases, respectively [22]. Equation (10) is purely kinematic in origin: wherever $w\rho$ has a gradient, there are more particles pointing (i.e., moving) down this gradient than up it, causing nonzero $\mathbf{P}$ [26].

For active particles, the leading-order mechanical stress is, as for ALCs, caused by their exerting force dipoles on the solvent [1]. In general, we can write $\mathbf{\Sigma} = -\zeta_P\mathbf{P}\otimes\mathbf{P}/\rho - \zeta_Q\mathbf{Q}$ with activity parameters ζ_P and ζ_Q. However, from Eq. (11), it follows that, if w is a function of ρ only (as assumed to this order in MIPS theories [20]), $\nabla\cdot\mathbf{Q}$ is a pure pressure gradient and, thus, ignorable. Expanding Eq. (10) as $\mathbf{P} = -\tau(w\rho)'\nabla\rho$ with prime denoting a ρ derivative, we recover Eq. (8), where

$$\hat{\kappa}(\phi) = \frac{\zeta_P}{\rho}\left(\frac{\tau(w\rho)'}{\phi'}\right)^2. \tag{12}$$

The sign of $\hat{\kappa}$ then depends only on whether swimmers are extensile ($\zeta_P > 0$) or contractile ($\zeta_P < 0$) [42]. Time-reversal interchanges these two cases, so the active contribution to $\hat{\kappa}$ in Eq. (8) is odd under it, unlike any passive part [45].

In keeping with our earlier discussion of Eq. (8), we now suppress the ϕ dependence of $\hat{\kappa}$. However, one could, alternatively, retain $\hat{\kappa}(\phi)$ in Eq. (12) as part of a "best-fit" procedure to a more detailed kinetic theory of MIPS [30]; we have checked numerically that the results are broadly similar to those with constant $\hat{\kappa}$ reported in what follows.

Next, we assume that local diffusive relaxation normal to interfaces ensures that their local structure is only weakly perturbed by fluid motion, just as applies in passive Model H [28]. We, thereby, nonperturbatively recover Eq. (9), now with $\tilde{\sigma} = \sigma\hat{\kappa}/\kappa$ [30]. So long as swimmers are extensile, the two interfacial tensions in Eq. (9) are positive, and the diffusive [$L \propto (\sigma t)^{1/3}$] and VH ($L \propto \tilde{\sigma} t/\eta$) regimes both remain intact. The prefactor of the VH coarsening is, however, shifted (possibly strongly), as is the crossover length $L_\times \propto (\eta\sigma/\tilde{\sigma})^{1/2}$.

In contrast, very different physics now arises for contractile swimmers. Here, the interfacial tension σ in the diffusive sector is positive as usual, but the mechanical one $\tilde{\sigma}$ is negative. Equation (9) still makes sense, but, instead of a crossover from diffusive to VH coarsening, there is a balance point $L_B \propto (\eta\sigma/|\tilde{\sigma}|)^{1/2}$ where diffusive coarsening is negated by VH anticoarsening. Thus, we predict that domain growth will cease at this scale, to be replaced by a dynamic equilibrium where the diffusive shrinkage of interfacial area is balanced by its production under the action of the contractile stress. This stretching of interfaces can be understood as a combination of the kinematics—causing swimmers at an interface to be aligned normal to it on average—with the flow pattern around a contractile swimmer, which pulls fluid inwards axially and expels it around the equator. For a swimmer normal to the interface (with either polarity) the latter is equivalent to a negative interfacial tension (see Fig. 1). This argument also shows why any ϕ dependence of $\hat{\kappa}$ (unless it changes sign) should not alter things qualitatively. Negative active tension has also been suggested in [39] by an argument that appears unrelated, since it applies equally to contractile and extensile cases.

To test these ideas, we have solved Active Model H numerically on a lattice of size 256×256 by a noise-free (i.e., $\mathbf{\Lambda} = \mathbf{0}$) hybrid Lattice Boltzmann scheme [47]. The system was initialized in a mixed state with uniformly distributed noise with $-0.1 < \phi(\mathbf{r}, 0) < 0.1$. We set $a = -b = -0.004$, $\kappa = 0.006$, $\lambda = 0$, and $\eta = 1.67$, varied $\hat{\kappa}$, and checked that setting $\lambda \neq 0$ does not qualitatively affect our conclusions.

These simulations confirm our expectation that contractile (but not extensile) activity should lead to the arrest of MIPS (see [30] for movies). This can be seen in Fig. 2 [see [30,40] for the definition of $L(t)$]. Snapshots of the dynamics in an arrested state are shown in Fig. 3; although the length scale is now fixed, the structure is highly dynamic [48]. As $\hat{\kappa}$ increases towards zero, the saturation length L_B grows, and for $-10^{-4} < \hat{\kappa} < 0$, coarsening does not arrest within the time window of our simulations.

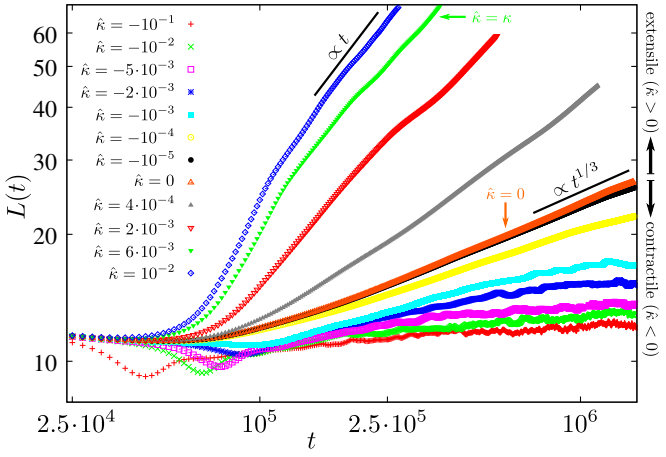

FIG. 2 (color online). Results for the domain size $L(t)$ in extensile ($\hat{\kappa} > 0$) and contractile ($\hat{\kappa} < 0$) two-dimensional scalar active fluids.

We speculate that it would do so eventually (at a length scale diverging as $\hat{\kappa} \to 0^-$), although our data do not rule out a finite negative threshold above which arrest never occurs. The saturation length was argued above to obey $L_B \propto (\eta\sigma/|\tilde{\sigma}|)^{1/2}$. At fixed η and κ this gives $L_B \propto |\hat{\kappa}|^{-1/2}$. Our data for $L_B(\hat{\kappa})$, instead, approach a weaker power law at low activity and a plateau at larger values (see Fig. 1 in [30]). The cause of this discrepancy is unclear but might suggest that a force balance different from Eq. (9) prevails in the highly dynamic states observed in strongly contractile systems.

The case $\hat{\kappa} = 0$ equates to passive Model B (no coupling to fluid motion), whereas passive Model H is attained for $\hat{\kappa} = \kappa$. The $L(t)$ curve is as expected in each case with final power laws close to $t^{1/3}$ and t, respectively. For passive Model H, the crossover in $L(t)$ from early-time diffusion to late-time fluid flow is sigmoidal, and the negatively curved part has often been interpreted as a sublinear power law [40]. Whatever the precise interpretation is, extensile systems with $0 < \hat{\kappa} < \kappa$ interpolate smoothly between the two pseudopassive limiting cases. Sigmoidal curves continue to be seen for systems with $\hat{\kappa} > \kappa$, in which the tension in the fluid sector is higher than the one driving diffusion. The data do not exclude an eventual power that exceeds unity but more likely reflect a prolonged sigmoidal crossover between diffusion and a linear growth with a large slope $\dot{L} \propto \tilde{\sigma}/\eta$.

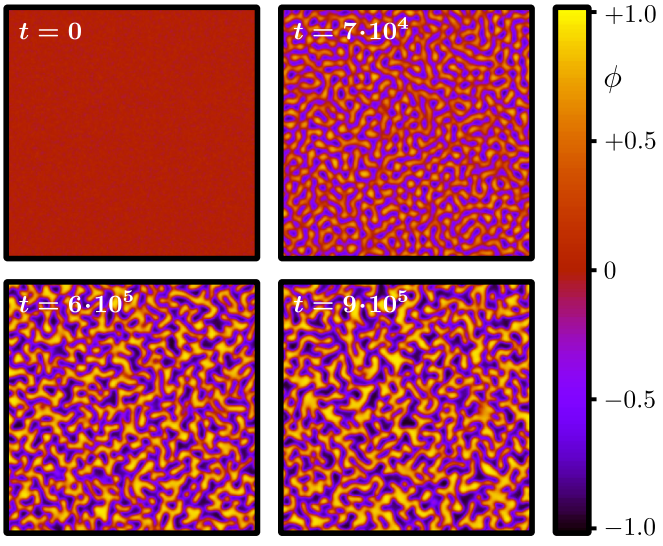

FIG. 3 (color online). Time series showing growth and arrest of domain structure in a two-dimensional contractile scalar active fluid with $\hat{\kappa} = -0.001$ and box size 256×256.

In summary, we have constructed Active Model H, a minimal model for scalar active matter coupled to a momentum-conserving solvent, by combining the traditional derivation of passive Model H, which has been fully tested experimentally, with a leading-order expression for the deviatoric stress. TRS is broken by two effects. One, encoded by λ, causes shifts in the densities at which phases can coexist in diffusive equilibrium [22]. The second effect is new, and amounts to a mismatch between the interfacial tension $\tilde{\sigma}$ that drives fluid motion and the tension σ that drives diffusive fluxes. For extensile particles (or weakly contractile ones if phase separation is driven by attractive interactions and not purely motility induced), both tensions are positive, and while their inequality violates TRS, its effect on coarsening dynamics is expected to be not qualitative. The opposite is true for contractile particles which become aligned normal to the interface between phases, creating a flow pattern that stretches it ($\tilde{\sigma} < 0$). Balanced by diffusion, this effect can cause the domain size $L(t)$ to saturate—consistent with data reported in [24]. This finding should also apply to active particles that are not entirely devoid of alignment interactions, as long as the system is in a parameter regime that avoids the formation of long-range orientational order.

Finally, it is tempting to associate our prediction of arrest at a finite length scale L_B in wet contractile scalar active fluids with observations of finite cluster formation, rather than full phase separation, in synthetic microswimmers undergoing MIPS [6]. However, there are two objections to this. First, we do not know which, if any, of these systems are contractile. Second, most observations of cluster phases are in "nearly dry" systems: clusters are found within two-dimensional layers close to a momentum-absorbing boundary [6]. Nonetheless, our prediction is that arrested separation should be generic in contractile wet systems undergoing MIPS. We look forward to future experimental tests of this prediction.

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[33] The parameter $\hat{\kappa} = \hat{\kappa}_R + \hat{\kappa}_D$ now contains a reversible contribution $\hat{\kappa}_R$ (even under time reversal) and a dissipative one $\hat{\kappa}_D$ (odd under time reversal) that stems solely from activity. If linear irreversible thermodynamics holds, we have $\hat{\kappa}_R = \kappa$. This is, however, not the case for systems sufficiently far from equilibrium [34].

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