



HAL
open science

An advected - field approach to the dynamics of fluid interfaces

Thierry Biben, Chaouqi Misbah, Anne Leyrat, Claude Verdier

► **To cite this version:**

Thierry Biben, Chaouqi Misbah, Anne Leyrat, Claude Verdier. An advected - field approach to the dynamics of fluid interfaces. EPL - Europhysics Letters, 2003, 63 (4), pp.623-629. 10.1209/epl/i2003-00564-y . hal-00197585

HAL Id: hal-00197585

<https://hal.science/hal-00197585v1>

Submitted on 15 Dec 2007

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

An advected - field approach to the dynamics of fluid interfaces

T. BIBEN AND C. MISBAH¹; A. LEYRAT AND C. VERDIER²

¹ *LSP-Grephe, CNRS/UJF - Université Joseph Fourier (Grenoble I) B.P.87, Saint Martin d'Hères, 38402 cedex France.*

² *Laboratoire de Rhéologie, CNRS/UJF/INPG - Université Joseph Fourier, Institut National Polytechnique de Grenoble and CNRS (UMR 5520), B.P. 53, Domaine Universitaire, 38041 Grenoble cedex 9, France.*

PACS. 82.70.Kj – Emulsions and suspensions.

PACS. 47.11.+j – Computational methods in fluid dynamics.

PACS. 47.55.Gf – Multiphase and particle-laden flows.

Abstract. – We present an Advected Field model of fluid interfaces at low Reynolds numbers and apply it to the fundamental problem of drop dynamics in an external flow. We focus on the ability of the numerical method to account for topology changes such as breakup, and compare the results obtained with this method to experimental data and theoretical predictions.

Introduction. – Since the pioneering experimental and theoretical work of Taylor [1, 2], the problem of droplet breakup, in view of its industrial relevance, has motivated a huge number of studies in various scientific communities like general physics, fluid mechanics, rheology and computational physics. The challenge is to understand and therefore give means to control the size of the fragments that are produced after breakup in out of equilibrium regimes. Using roller devices, Taylor was able to produce two-dimensional shear and elongational flows, and applied them to investigate droplet deformation in a suspending fluid. The relevant dimensionless parameters for this problem are the capillary number $Ca = \eta_{out} R \dot{\gamma} / \Sigma$ (where η_{out} is the suspending fluid viscosity, R is the initial droplet radius, $\dot{\gamma}$ is the deformation rate and Σ is the interfacial tension), which is related to the droplet deformability, and the viscosity ratio $p = \eta_{in} / \eta_{out}$ (η_{in} is the viscosity of the drop), as demonstrated experimentally by Grace [3]. Critical capillary numbers for breakup are indeed determined as functions of p , in shear and elongational flows over ten decades, as well as for mixed flows [4]. Analytical models [5, 6] based on the perturbation theory have been developed since, as well as numerical models, which are more appropriate for large deformations [7] [8–11]. While the latter are based on the boundary integral method [8–11], the level set method [12] has been introduced recently to cope with several difficulties encountered in the boundary integral formulation, like the front tracking problem and the treatment of breakup itself. The key point in the level set method is to regularize the macroscopically infinitely thin interface by introducing a smoothing function which is constructed from the distance field to the interface. Applications

to binary fluids have been considered very recently [13,14]. Similar methods like the volume-of-fluid method have been used as well (see [15]). In the same spirit, Phase-Field models were introduced twenty years ago in the field of crystal growth, with the major difference that the auxiliary phase field has a physical meaning: it accounts for the solid-liquid phase equilibrium, and is then constructed from thermodynamical considerations. These Phase-Field methods should then naturally apply to binary fluids, with the same accuracy as level set methods, but should incorporate more physical ingredients, such as short range interactions between the interfaces (correlations due to the broad microscopic nature of the interfaces) which are absent in level set approaches. Indeed, Loewenberg and Hinch [16] have shown that in the absence of short range attractions (like the Van der Waals force) coalescence cannot be accounted for, which renders the determination of the rheological properties of concentrated emulsions difficult. Phase Field models incorporate the short range physics of the interface naturally. Coalescence can occur due to the short range interactions between the interfaces and breakup is controlled by the free energy functional of the Phase-Field itself. Van der Waals forces can be added to account for polarizability effects when necessary. Although Phase Field approaches usually give a good description of the physical phenomena (see for example [17] and references therein), they often fail to provide quantitative results since in practice the broadness of the interface is overestimated. The scope of this letter is to present a modified Phase Field approach which allows for quantitative predictions. This method should be numerically more efficient than traditional boundary integral formulations for concentrated emulsions since the resolution time mainly depends on the box volume which scales like the number N_d of droplets, at a given concentration, whereas boundary integral methods scale like N_d^2 in the most favorable situation (no viscosity contrast). We will however focus here on the ability of the method to give quantitative predictions for a single droplet, where many results are available, which is certainly the worst situation from the computational point of view in comparison to other methods.

In a first part we discuss the Phase-Field formulation for binary fluids, and show that slight modifications are necessary to account for the proper macroscopic physics. As a result, the new method is intermediate between the pure Phase-Field formulation and the level-set method. We will now use the name "Advection-Field" method (AF) for this modified formulation, which will be justified later. Comparison of the predictions of the AF approach with available experimental and theoretical data for the breakup problem are presented in the second part.

The advected field approach. – A binary fluid can be described by an order parameter field which identifies the two components. Using a Landau-Ginzburg description, a purely isotropic fluid can be described by a scalar order parameter field $\phi(\mathbf{r}, t)$ where \mathbf{r} denotes the spatial coordinates while t is time. The simplest free energy functional that can be written to describe a binary coexistence is the usual square gradient theory:

$$\mathcal{F}[\phi] = \int_{Volume} \left\{ W(\phi) + \frac{\epsilon^2}{2} [\nabla\phi]^2 \right\} d\mathbf{r} \quad (1)$$

where $W(\phi)$ is a double-well potential describing the bulk coexistence of the two phases, and the square gradient contribution sets the width of the interface to $\sqrt{2}\epsilon$. A simple prescription for $W(\phi)$ is $W(\phi) = (1 - \phi^2)^2/4$ which determines the coexisting values of the field, ± 1 . In a Cahn-Hilliard spirit, a dynamical equation can be written for the temporal evolution of the order parameter field which is advected (*i.e.* transported) by the flow:

$$\frac{\partial\phi}{\partial t} + \mathbf{v} \cdot \nabla\phi = -\Gamma \left(\frac{dW}{d\phi} - \epsilon^2 \Delta\phi \right) \quad (2)$$

where \mathbf{v} is the macroscopic velocity field. While the left contribution expresses the hydrodynamic transport of the field by the flow, the right term is responsible for the chemical (diffusive) relaxation of the field to its equilibrium value. Γ is a constant which sets the relaxation time of this process. A conservative dynamics (*i.e.* $\int \phi d\mathbf{r} = cste$) can be built by substituting $-\Gamma\Delta$ to Γ . This Cahn-Hilliard formulation (2), very similar to the formulation already given by Jasnow and Viñals [17], cannot be used however to describe the dynamics of a mesoscopic droplet (radius $R \sim 100\mu m$) in a flow. A first practical reason is due to the value of ϵ which should be of the order of $10^{-4}R$ to describe the actual width of a binary interface. This value is out of reach at the moment for present computers and a typical accessible value is $10^{-2}R$ ($0.04R$ in the present work). The second and more serious problem comes from the free energy formulation itself. The velocity field appearing in (2) is the macroscopic velocity field, which expresses the transport of matter. The velocity of the interface must then correspond to the local velocity field. This is not indeed the case with the above formulation, which can be illustrated quite simply: if we cancel the velocity field in (2), and start from an initial arbitrary shape for the drop (*i.e.* the zero isocontour of the order parameter field ϕ), according to the velocity field the shape should not change. Solving (2) however will lead to a spherical shape if the dynamics is conserved or the drop will simply disappear if the dynamics is not conserved, which simply expresses the minimization of the free energy functional (1). The basic reason for this problem is due to surface tension of the order parameter field, which is not coupled to the velocity field, and thus induces a relaxation even in the absence of a flow. A simple prescription which cancels this effect is to add the counter term suggested by Folch et al. [18] in their study of the Saffman-Taylor instability, and to solve the following equation:

$$\frac{\partial \phi}{\partial t} + \mathbf{v} \cdot \nabla \phi = -\Gamma \left(\frac{dW}{d\phi} - \epsilon^2 [\Delta \phi + c |\nabla \phi|] \right) \quad (3)$$

where " c ", appearing in the counter term, is the local curvature field defined everywhere in space, which corresponds to the curvature of the isocontours of ϕ . The precise expression of this counter term can be justified by an analysis of the asymptotic behaviour of this formulation when ϵ goes to zero (the "sharp interface limit" in the Phase-Field terminology). We cannot reproduce this detailed analysis here, but this term simply cancels to first order in ϵ the curvature dependence of the interfacial hyperbolic tangent equilibrium profile of the interface, $\phi(r) = \tanh(r/\sqrt{2}\epsilon)$, where r is the algebraic distance to the $\phi = 0$ isocontour. In other words, the laplacian appearing in (2) is replaced by $\partial^2 \phi / \partial r^2$ and thus the Cahn Hilliard prescription only plays a role in the normal direction. The interface is thus treated as locally flat by the microscopic Cahn Hilliard theory. The global shape evolution of the interface (*i.e.* in the parallel direction) is entirely contained in the advection term, and is thus controlled by the Stokes equation alone. The normal and the parallel directions have thus been decoupled, keeping the physical ingredients of the Cahn Hilliard theory for a microscopically quasi-planar interface in the normal direction, and removing the unphysical coupling with the global macroscopic shape relaxation induced by a strong overestimation of the interfacial width due to numerical considerations. With this formulation, the order parameter field ϕ becomes a quasi-passive field (*i.e.* the $\phi = 0$ isocontour is transported by the flow), the Advected Field, but the shape of the interface is fixed to its hyperbolic tangent shape thanks to the new restoring term (r.h.s. of (3)).

Obviously, surface tension is a fundamental physical ingredient for the description of droplets dynamics, and it has to be incorporated in the dynamical equation for the velocity field:

$$\rho \left(\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} \right) = \nabla \cdot \bar{\bar{\sigma}} - \nabla P + \mathbf{F}_{interface} \quad (4)$$

where ρ is the density of the fluid, $\overline{\sigma}$ is the extra stress tensor of the fluid, P is the local pressure and $\mathbf{F}_{interface}$ is the force exerted by the interface on the fluid. This last term can be expressed by $\mathbf{F}_{interface} = d c \Sigma \delta(r) \hat{\mathbf{n}}$, where d is the interface dimensionality, c is the mean curvature field introduced above, Σ is the actual interfacial tension, r is the algebraic distance to the interface (the $\phi \equiv 0$ isosurface) and $\hat{\mathbf{n}}$ is the normal outgoing vector at the interface. We used the convention that $c = -\nabla \cdot \hat{\mathbf{n}}/d$, which leads to a negative value for a sphere. $\delta(r)$ is the Dirac distribution in the sharp interface limit, which can be regularized at finite values of ϵ by setting $\delta(r) \hat{\mathbf{n}} \rightarrow \nabla \phi/2$, where we have used the convention that $\phi < 0$ inside the drop. Since the AF goes from -1 to $+1$ while crossing the interface, we can easily check the normalization condition whatever the shape of the profile. The final expression for $\mathbf{F}_{interface}$ is then:

$$\mathbf{F}_{interface} = \frac{d}{2} c \Sigma \nabla \phi \quad (5)$$

$\overline{\sigma}$, the stress tensor, is chosen to obey the local Newtonian law : $\overline{\sigma}_{ij} = \eta(\phi) (\partial_i v_j + \partial_j v_i)$. We have emphasized in this expression the spatial dependence of the viscosity η which accounts for a viscosity contrast between the internal fluid (i.e. the drop) and the external medium. The simple prescription $\eta = \eta_{in}(1 - \phi)/2 + \eta_{out}(1 + \phi)/2$ allows the viscosity to vary smoothly across the interface to interpolate between the internal value η_{in} and the external viscosity η_{out} . Note that the method can also allow for a density contrast, but it is not the purpose here. Since we consider droplets of typical size about $100 \mu m$, viscosities around $1 Pa.s$ or more, and rates in the range $[10^{-2} - 10^2 s^{-1}]$, the Reynolds number $Re = \rho a^2 \gamma / \eta_{out}$ should be smaller than 10^{-3} and inertial effects are negligible, the momentum equation then simply reduces to the Stokes equation where ρ has been set to zero. The pressure field can easily be eliminated in Fourier space thanks to the incompressibility condition $\nabla \cdot \mathbf{v} = 0$. The solution of these dynamical equations can be obtained numerically by solving the AF equation (3) in the direct space using a finite difference method while the Stokes equation (which we choose to solve using a relaxation scheme, keeping $\rho \partial \mathbf{v} / \partial t$) is best solved in the Fourier space, using an implicit scheme. For this purpose we used a regular mesh with a square or cubic unit cell, depending on dimensionality, and took special care of anisotropy problems due to the discretization of the differential operators. We used in practice operators which are isotropic up to order three in the lattice spacing. The lattice spacing is chosen to be exactly ϵ , whose value is fixed to $0.04R$, where R is the radius of the drop at rest. When an external flow is imposed, the total flow can, without any restriction, be separated into two components $\mathbf{v} = \mathbf{v}_{ext} + \mathbf{u}$ where \mathbf{v}_{ext} is the applied external flow, a shear or an elongational flow, and \mathbf{u} is the flow induced by the droplet. The boundary conditions at the edges of the mesh have been chosen to be periodic for the AF as well as the component of the flow \mathbf{u} , \mathbf{v}_{ext} being treated analytically.

Results. – To illustrate the ability of the method to account for the breakup process we show in Fig.1 a breakup sequence in 3D for an initially elongated drop (spherocylindrical geometry) relaxing in a quiescent fluid. To test quantitatively the method, we compared the data provided by the AF method to the analytical study made in 2D by Buckmaster and Flaherty [6] for a drop placed in an elongational flow defined by: $v_{ext}^x = \gamma x$ and $v_{ext}^y = -\gamma y$, where x is the coordinate along the elongation axis. Interestingly these authors were able to calculate the deformation of a drop in an elongational flow up to the burst transition. The two dimensionless relevant parameters are the viscosity ratio p and the capillary number Ca as defined in the introduction. Below a certain critical value Ca_c the drop deforms under the elongational constraint to reach a steady shape, which can be characterized by its “elongation” $1 - B/L$ where B is the width and L is the length. Above Ca_c no stationary shape exists and

3D

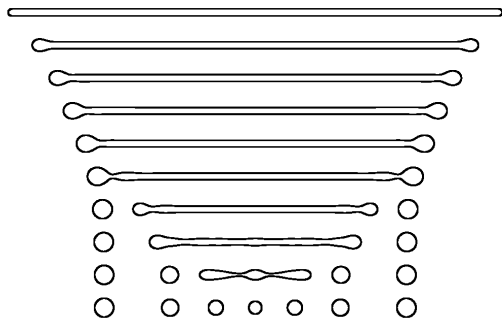


Fig. 1

Fig. 1 – Breakup sequence of an initially elongated droplet in 3D for $p = 1$, the AF method allows to follow the full dynamics of the breakup. The grid size is 800x200.

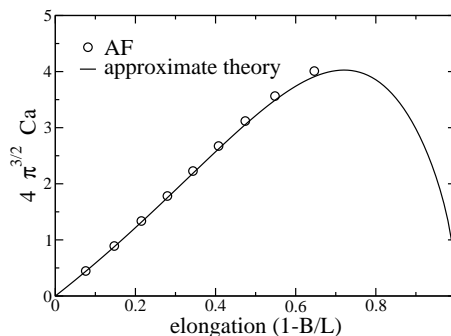


Fig. 2

Fig. 2 – Comparison between the AF method and the theoretical predictions by Buckmaster and Flaherty in 2D for drop deformation in an elongational flow. The grid size is 300x300

the drop elongates until it breaks. Such a dynamic behaviour corresponds to a saddle-node bifurcation, and thus an unstable branch should exist that merges with the stable branch at the critical capillary number. This is precisely the scenario found by Buckmaster and Flaherty [6]. This type of bifurcation seems to be quite generic in the physics of deformable objects under hydrodynamic constraint, it also occurs for vesicles (tank-treading to flipping transition) [20].

We can see in Fig.2 a comparison of the AF method with the analytical theory of [6]. This figure shows the predicted elongation of the drop at a given value of the capillary number for a viscosity ratio $p = 1$ in 2D. At a given value of Ca two shapes satisfy the stationary condition below the critical capillary number $Ca_c \sim 0.18$ (or equivalently $4\pi^{3/2}Ca \sim 4$ from Fig.2): convex shapes when $1-B/L < 0.72$ and non-convex shapes above 0.72, and no stationary shape exists above Ca_c . While the less elongated shape corresponds to the actual stationary shape, the most elongated one is unstable and thus is not physical. The AF model seems to provide quite good quantitative agreement with the analytical theory, even close to the critical capillary number since the approximation presented in Fig.2 deviates from the full theory [6]. The value of the critical capillary number $Ca_c = 0.18$ obtained with the AF method agrees perfectly well with the theoretical expectation.

The 3D situation can be investigated as well. It allows for a more precise comparison with experimental data. In Fig.3 we present the results obtained for the critical capillary number as a function of the viscosity ratio in the particular case of an axisymmetric geometry, which allows for a reduction of the problem to a two-dimensional resolution. One must note that the 3D axisymmetric elongational flow is defined by: $v_{ext}^x = \gamma x$ and $v_{ext}^r = -\frac{\gamma}{2} r$, where r denotes the radial coordinate normal to the elongation axis x . This differs from the usual experimental situation corresponding to the four roll apparatus [4] which indeed corresponds to a two-dimensional elongational flow. The pure 2D AF resolution is indicated in Fig.2 for comparison. The dimensionality seems to have a strong influence on the results, although we can note a good qualitative overall agreement between models and experimental findings.

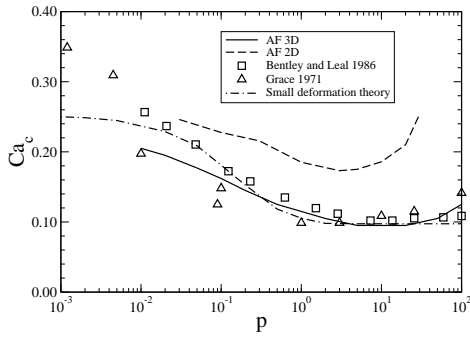


Fig. 3

Fig. 3 – Critical capillary number as a function of the viscosity ratio for an elongational flow. Squares and triangles correspond to experimental data while the continuous line is the AF 3D prediction, and the dashed line is the 2D prediction. The small deformation theory corresponds to the dot-dashed line. The grid size is 300x300

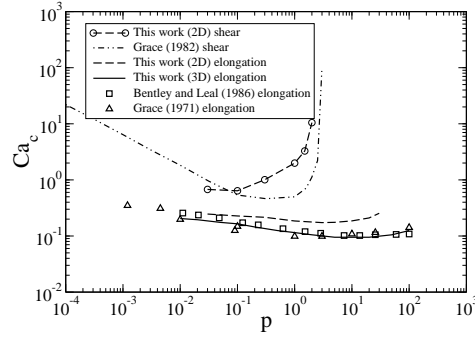


Fig. 4

Fig. 4 – Same figure for elongational (bottom curves) and shear (top curves) flows. Squares and triangles are experimental data for elongational flow, while the AF predictions are the straight line in 3D and the dashed line in 2D. The dot-dashed line indicates the average location of experimental data for shear flows, while the dashed line with circles corresponds to the AF prediction in 2D.

Interestingly, the largest deviations are observed in the low viscosity ratio regime, where both the small deformation theory and the AF method fail to reproduce the observed growth of the critical capillary number. This failure seems to be due to the particular shape of the droplets in these extreme regimes. Indeed, as can be seen in Taylor's paper [2], droplets tends to develop pointed ends, that can hardly be accounted for by a small deformation theory, but also by the AF approach. This failure of the AF approach is due to the width ϵ of the interface, which is a natural cutoff for the local curvature radius. Pointed ends cannot be observed in such situations, rather, the drop shape is seen to converge to a limiting shape when the viscosity ratio p decreases to zero. However, this problem only occurs for very low values of the viscosity ratio $p < 0.01$, and depends strongly on the choice of ϵ . This problem does not occur in the high viscosity ratio regime since shapes are relatively ellipsoidal in this regime, so that the curvature radius is always large compared to ϵ .

In Fig.4 we show a comparison (log-log scale) between 2D-AF results obtained for a pure shear flow corresponding to $v_{ext}^x = \gamma y$ and $v_{ext}^y = 0$ with experimental data. Also shown are the AF results (2D and 3D) for elongational flows. A comparison between the 2D AF results for a shear flow and experimental data can only be qualitative as results from the previous discussion, but we can observe the same trend in the large viscosity ratio regime: for p larger than 3.5 droplets cannot be broken anymore. The shear situation differs then significantly from the pure elongational case. In fact, a shear flow can be separated into two components, a purely elongational flow which axes are oriented at 45 degrees from the shear direction, and a purely rotational flow. While the elongational component tends to elongate the drop, the rotational component deflects it from the main elongation axis. Since the rotational torque felt by the drop increases with the viscosity ratio (a purely rigid ellipsoid would have a flipping motion in a shear flow [19]) elongation becomes very inefficient, and breakup does not occur

anymore. This trend is perfectly visible in Fig.4 for both the 2D AF model and experimental results. Full 3D simulations in the shear case should give better quantitative agreements with experimental data, as it is the case in the elongation situation, but although the numerical implementation is not sensitive to dimensionality, the computational time scales like N^d where N is the number of grid points in a given direction and d the dimensionality. A systematic study then requires much more computational time and is not presented here.

Conclusion. – Apart from the very specific regime where pointed ends can occur (very low viscosity ratios) , all these results show that the AF model captures perfectly well the physical features involved in the drop deformation problem. First the burst transition which separates the existence of stationary shapes from the unsteady regime where the drop elongates is well described by the numerical model, as shown in 2D in Fig.2. Moreover the method is able to account for topological changes and gives access to the full dynamics of the breakup process. Comparison with experimental data gives a fair agreement, in view of the scattering of experimental data and the dimensionality difference and sensitivity. We considered here a single droplet submitted to an external flow, and the same model can be used without modification to model a set of droplets (only the initial condition needs to be changed). It must be noted that this method also captures the dynamics of the fusion process, as will be shown elsewhere, since the short range attraction induced between the interfaces by the AF method is able to overcome the hydrodynamic repulsion, which is a problem in pure hydrodynamic approaches. Unlike integral formulations, which explicitly take advantage of the linearity of the Stokes equation for Newtonian systems, the AF method can be used to model non-Newtonian fluids, and we believe that this method will be helpful to model problems like the dynamics of multiphase flows. We must also mention that the AF method is not restricted to binary systems, the AF method has already been applied to investigate the dynamics of more complex interfaces such as phospholipid membranes. An application to vesicle dynamics can be found in [20].

REFERENCES

- [1] G.I. Taylor, Proc. Royal Soc., **A138**, 41 (1932).
- [2] G.I. Taylor, Proc. Royal Soc., **A146**, 501 (1934).
- [3] H.P. Grace, Chem. Eng. Commun., **14**, 225 (1982).
- [4] B.J. Bentley and L.G. Leal, J. Fluid Mech., **167**, 241 (1986).
- [5] D. Barthès-Biesel and A. Acrivos, J. Fluid Mech., **61**, 1 (1973).
- [6] J.D. Buckmaster and J.E. Flaherty, J. Fluid Mech., **60**, 625 (1973).
- [7] J.M. Rallison and A. Acrivos, J. Fluid Mech., **89**, 191 (1978).
- [8] H.A. Stone and L.G. Leal, J. Fluid Mech., **198**, 399 (1989).
- [9] H.A. Stone and L.G. Leal, J. Fluid Mech., **206**, 223 (1989).
- [10] J.M. Rallison, Ann. Rev. Fluid Mech., **16**, 45 (1984).
- [11] H.A. Stone, Ann. Rev. Fluid Mech., **26**, 65 (1994).
- [12] M. Sussman, E. Fatemi, P. Smereka, S. Osher, Computers and Fluids, **27**, 663 (1998).
- [13] Y.Y. Renardy and V. Cristini, Phys. of Fluids, **13**, 2161 (2001).
- [14] S.B. Pillapakam and P. Singh, J. Comput. Physics, **174**, 552 (2001).
- [15] J. Li, Y. Y. Renardy and M. Renardy, Phys. Fluid, **12**, 269 (2000).
- [16] M. Loewenberg and E. J. Hinch, J. Fluid Mech. , **338**, 299 (1997).
- [17] D. Jasnow and J. Viñals, Phys. Fluids, **8**, 660 (1996).
- [18] R. Folch, J. Casademunt, A. Hernández-Machado and L. Ramírez-Piscina, Phys. Rev. E, **60**, 1724 (1999).
- [19] G. B. Jeffery, Proc. R. Soc. Lond. A **102**, 161 (1922).
- [20] T. Biben and C. Misbah, Physical Review E **67**, 031908 (2003).