Bénard-Marangoni instability in a linear viscoelastic magnetic fluid

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ABSTRACT

In this manuscript we report theoretical and numerical results on the convection of a magnetic fluid with a viscoelastic carrier liquid. The viscoelastic properties are given by the linear Oldroyd model or equivalently by linear relaxing elasticity. We take the lower interface to be rigid, whereas the upper, free one is assumed to be non-deformable and flat. At that interface the surface tension varies linearly with the temperature. Using a spectral method we calculate numerically the convective thresholds for both, stationary and oscillatory bifurcation. The effects of viscoelasticity and of a magnetic field on the instability thresholds are emphasized.

INTRODUCTION

Ferrofluids are magnetic fluids formed by a stable colloidal suspension of magnetic nanoparticles dispersed in a carrier liquid. Without an applied external magnetic field the orientations of the magnetic moments of the particles are random resulting in a vanishing macroscopic magnetization. An external magnetic field, however, easily orients the particles' magnetic moments and a large (induced) magnetization is obtained. There are two main features that distinguish ferrofluids from ordinary fluids, the polarization force and the body couple[1]. In addition, when a magnetic field is applied, the ferrofluid can exhibit additional rheological properties such as magneto-viscosity, adhesion properties, and non-Newtonian behavior, among others[2]. In the last decades much efforts have been dedicated to the study of convection mechanisms in ferrofluids. In addition, heat transfer through magnetic fluids, in particular, has been one of the leading areas of scientific study due to its technological applications[3].

The first continuum description of magnetic fluids was given by Neuringer and Rosensweig[4]. Later, Finlayson [5] studied the convective instability of a magnetic fluid for a fluid layer heated from below in the presence of a uniform vertical magnetic field. He discussed the cases of both, shear free and rigid horizontal boundaries within the linear stability method. Ryskin and Pleiner[6], using nonequilibrium thermodynamics, have derived a complete set of equations to describe ferrofluids in an external magnetic field. Recently, the thermal convection in viscoelastic magnetic fluids was studied for idealized and rigid boundary conditions[7,8].

On the other hand, the Marangoni instability is a good example of a surface tension driven instability. If a temperature gradient is applied to a layer of a fluid with a free surface, the heat conducting state becomes unstable, and convection starts above a critical temperature gradient, when the heating is done from below. The linear analysis of the convection in a magnetic fluid with deformable free surface was studied by Weilepp and Brand[9] and by Hennenberg *et al.*[10]. The linear and weakly nonlinear analysis in the case of viscoelastic pure fluids was performed for a non-deformable free surface by Lebon *et al.*[11,12]. The eigenvalues and eigenfunctions of the adjoint problem and adjoint boundary conditions for the case of a deformable free surface for the Marangoni problem have been derived only recently[13].

The purpose of the present work is to analyze the influence of the viscoelasticity on Bénard-Marangoni convective thresholds in a magnetic fluid; in particular, where the separation ratio and magnetic separation ratio are not too large, the simple fluid approximation can be used[6]. To this aim an Oldroyd viscoelastic magnetic fluid heated from below is considered. The description of the system involves many parameters whose values have not yet been determined accurately. Therefore, we are left with some freedom in fixing the parameter values. Since the boundary conditions are complicated, we numerically solve the linearized system using a collocation spectral method in order to determine the eigenfunctions and eigenvalues and consequently the convective thresholds. The paper is organized as follows: In the following section the basic hydrodynamic equations for viscoelastic magnetic fluid convection are presented, followed by the linear stability analysis, and, finally, conclusions are presented.

THEORETICAL MODEL

Let us consider a layer of thickness d of an incompressible, magnetic fluid in a viscoelastic carrier liquid, with very large horizontal extension (in the xy-plane) in the vertical gravitational field $\mathbf{g}$ and subject to a vertical temperature gradient. The magnetic fluid properties can be modeled as of electrically nonconducting superparamagnets. The magnetic field $\mathbf{H}$ is assumed to be parallel to the z axis, $\mathbf{H} = H_0\,\hat{\mathbf{z}}$. It would be homogeneous, if the magnetic fluid were absent. Let us choose the z-axis such that $\mathbf{g} = -g\,\hat{\mathbf{z}}$ and that the layer has its interfaces at coordinates $z = -d/2$ and $z = d/2$. A static temperature difference across the layer is imposed, $T(z = -d/2) = T_0 + \triangle T$ and $T(z = d/2) = T_0$. We impose the lower interface to be rigid and the upper one free. The latter one is assumed to be non-deformable and flat, which is a reasonable approximation for very small capillarity numbers[11,12]. At the upper, free interface, the surface tension Σ is taken to vary linearly with the temperature. Under the Boussinesq approximation, the dimensionless linear perturbation equations read[8,13]

$$\boldsymbol{\nabla}\cdot\mathbf{v} = 0 \tag{1}$$

$$P^{-1}\partial_t\mathbf{v}+\boldsymbol{\nabla}p = \boldsymbol{\nabla}\cdot\overline{\boldsymbol{\tau}}+Ra\,\Pi_1(\theta,\phi)\hat{\mathbf{z}} \tag{2}$$

$$\partial_t U_{ij} = \overline{D}_{ij} - \Gamma_1^{-1}U_{ij} \tag{3}$$

$$\partial_t(\theta - M_4\partial_z\phi) = (1-M_4)w + \nabla^2\theta \tag{4}$$

$$(\partial_{zz} + M_3[\partial_{xx} + \partial_{yy}])\phi - \partial_z\theta = 0 \tag{5}$$

$$\nabla^2\phi_{ext} = 0 \tag{6}$$

with the dimensionless linearized viscoelastic stress tensor $\overline{\tau}_{ij} = -E_1 U_{ij} - \overline{D}_{ij}$, where p is the static hydrodynamic pressure and $\overline{D}_{ij} = \frac{1}{2}(\nabla_i v_j+\nabla_j v_i)$ is the so-called strain-rate tensor. The driving of the system is described by $\Pi_1(\theta,\phi) = (1+M_1)\theta - (M_1 - M_5)\partial_z\phi$.

The variables are $\{\mathbf{v}, U_{ij}, \theta, \phi\}$, the dimensionless velocity perturbation, the dimensionless strain tensor, the temperature perturbation and the dimensionless magnetic potential perturbation, respectively. In Eqs. (1) - (6), the following groups of dimensionless numbers have been introduced: ***(a)*** (pure fluids) The Rayleigh number, $Ra = \alpha_T g\beta d^4/\kappa\nu$, accounting for buoyancy effects; and the Prandtl number, $P = \nu/\kappa$, relating viscous and thermal diffusion time scales. ***(b)*** (magnetic fluid) The strength of the magnetic Kelvin force relative to buoyancy is measured by the parameter $M_1 = (\beta\chi_T^2/\rho_0 g\alpha_T)H_0^2/(1+\chi)$; the nonlinearity of the magnetization, $M_3 = 1 - (\chi_H H_0^2)/(1+\chi)$, a measure of the deviation of the magnetization curve from the linear behavior $M_0 =$

$\chi_0 H_0$; the relative strength of the temperature dependence of the magnetic susceptibility $M_4 = (\chi_T^2/c_H)H_0^2 T_0/(1+\chi)$; and the ratio of magnetic variation of density with respect to thermal buoyancy $M_5 = (\alpha_H/\alpha_T)\chi_T H_0^2/(1+\chi)$. ***(c)***(viscoelastic fluid) The elastic modulus $E_1 = K_1 d^2/\nu\kappa$ and the strain relaxation $\Gamma_1 = \nu\kappa\tau_1/d^2$.

In these dimensionless numbers different physical quantities appear such as ρ_0 the reference mass density, c_H the specific heat capacity at constant volume and magnetic field, T_0 the reference temperature, H_0 the reference magnetic field, χ_T the pyromagnetic coefficient, κ the thermal diffusivity, χ_H the longitudinal magnetic susceptibility, α_T the thermal expansion coefficients and α_H the magnetic expansion coefficients, ν the static viscosity, τ_1 the strain relaxation time, K_1 is the plateau elastic modulus, and $\beta = \triangle T/d$.

Instead of the linear viscoelastic equation in the form of a relaxing strain tensor, Eq.(3), often a heuristic constitutive equation, the linear Oldroyd model, is used

$$(1+\Gamma\partial_t)\overline{\boldsymbol{\tau}} = (1+\Lambda\partial_t)\overline{D}_{ij} \qquad (7)$$

containing the Deborah number, $\Gamma = \lambda_1\kappa/d^2$, with λ_1 the stress relaxation time, and the retardation number, $\Lambda = \lambda_2/\lambda_1$, the ratio between the strain rate relaxation, λ_2, and λ_1. Within the linear domain both descriptions are equivalent with $\Gamma = \Gamma_1$ and $\Lambda = (1+E_1\Gamma_1)^{-1}$, revealing however that Λ is restricted by $0 < \Lambda < 1$. The static viscosity ν, used to scale the time in the viscoelastic description, is related to the asymptotic viscosity ν_∞ (used in the Oldroyd case) by $\nu_\infty = \nu/\Lambda$ The main advantage of the use of the explicit viscoelasticity, Eq.(3), is that it can easily be generalized in a straightforward physical manner into the nonlinear domain. In addition, for complex liquids that need the introduction of additional degrees of freedom the combination of the strain tensor dynamic equation with those additional degrees of freedom follows standard thermodynamic and hydrodynamic procedures, while the heuristic generalization of the constitutive equation reaches its limits, rapidly. Furthermore, realistic boundary conditions are straightforward for the strain field, but not at all if the stress is used as variable. A critical comparison between the two approaches at the quadratic nonlinear level is given in Ref. 14. Since we will discuss only linear properties, we will use both descriptions in parallel.

Let us comment on the numerical values of the parameters; the Rayleigh number Ra can be changed by several orders of magnitude by varying the applied temperature gradient, with $Ra \sim 10^2 - 10^3$ relevant in the present case. A typical value for P in viscoelastic fluids is $P \sim 10^0 - 10^3$ with $Pr \sim 10$ for aqueous systems. The magnetic numbers are field dependent with $M_1 \sim 10^{-4} - 10$, $M_3 \gtrsim 1$, $M_4 \sim M_5 \sim 10^{-6}$ for typical magnetic field strengths.[6,7,8] M_1 is directly proportional to H_0^2, while M_3 is only a weak function of the external magnetic field. Since M_4 and M_5 are very small and not related to viscoelastic effects, which we are interested in here, we expect not to loose any reasonable aspect of the problem under consideration by putting them to zero. Kolodner[15] and the group of Chu[16,17,18] have suggested that for aqueous suspensions the Deborah number is about $\Gamma \sim 10^{-3} - 10^{-1}$, but for other kinds of viscoelastic fluids the Deborah number can be of the order of $\Gamma \sim 10^3$. Unfortunately, no experimental data are available for the retardation number nor for the stress relaxation time, so we take $\Lambda = 2$, where fixed.

LINEAR STABILITY ANALYSIS

In order to investigate the linear stability of the quiescent ground state under increasing Rayleigh and M_1 numbers, we first eliminate the effective pressure and two components of the velocity field by applying the curl ($\boldsymbol{\nabla} \times ...$) and double curl ($\boldsymbol{\nabla} \times \boldsymbol{\nabla} \times ...$) on the Navier-Stokes equation and then considering only w, the z-component of tvelocity. After some algebra, the linear equations read

$$P^{-1}\partial_t\nabla^2 w = \nabla^2(\boldsymbol{\nabla}\cdot\overline{\boldsymbol{\tau}})_z + Ra\nabla_\perp^2\mathcal{L}_\Sigma \quad (8)$$

$$(1+\Gamma\partial_t)(\boldsymbol{\nabla}\cdot\overline{\boldsymbol{\tau}})_z = (1+\Gamma\Lambda\partial_t)\nabla^2 w \quad (9)$$

$$\partial_t\theta = w + \nabla^2\theta \quad (10)$$

$$(\partial_{zz} + M_3\nabla_\perp^2)\phi - \partial_z\theta = 0 \quad (11)$$

with $\mathcal{L}_\Sigma = (1+M_1)\theta - M_1\partial_z\phi$. We remark that Eqs. (8) and (9) can be combined in order to eliminate the stress tensor (or the strain tensor) resulting in a single equation for w. Defining a vector field $\mathbf{u} = (\theta,\phi,w)^T$ that contains the important variables for the linear analysis, the spatial and temporal dependencies of $\mathbf{u}$ are separated using a normal mode expansion

$$\mathbf{u}(\mathbf{r},t) = \bar{\mathbf{U}}(z)\exp[i\,\mathbf{k}\cdot\mathbf{r}_\perp + s\,t], \quad (12)$$

with $\bar{\mathbf{U}} = (\Theta,\Phi,W)^T$, and where $\mathbf{k}$ is the horizontal wavenumber of the perturbations, $\mathbf{r}_\perp$ is the horizontal position vector and $s = \sigma + i\,\Omega$ denotes the complex eigenvalues with σ the linear growth rate of a periodic perturbation and Ω its frequency. Using the ansatz (12), Eqs. (8) - (11) are reduced to the following coupled ordinary differential equations

$$D^2\Theta = (k^2+s)\Theta - W \quad (13)$$

$$D^2\Phi = M_3k^2\Phi + D\,\Theta \quad (14)$$

$$D^4W = \xi_1D^2W - \xi_2W + \xi_3\Theta - \xi_4D\Phi \quad (15)$$

where $D^nf = \partial_z^nf$, $\xi_1 = 2k^2 + s\mathcal{Q}/P$, $\xi_2 = k^2(k^2+s\mathcal{Q}/P)$, $\xi_3 = Ra\,k^2\,(1+M_1)\,\mathcal{Q}$, and $\xi_4 = Ra\,k^2M_1\mathcal{Q}$ with $\mathcal{Q} = (1+s\Gamma)/(1+s\Lambda\Gamma)$.

The correct fluid and thermal boundary conditions[19] for a viscous or viscoelastic fluid are, at the lower rigid surface $(z=-1/2)$

$$\begin{aligned} &W = D\,W = 0 \\ &\Theta = 0, \end{aligned} \quad (16)$$

and at the upper free surface $(z=+1/2)$

$$\begin{aligned} &W = D^2W + \frac{k^2Ma}{\mathcal{Q}}\Theta = 0 \\ &(D+Bi)\,\Theta = 0, \end{aligned} \quad (17)$$

where $Ma = \gamma_T\beta d^2/\kappa\nu$ is the Marangoni number, which arises from the variation of the surface tension Σ with temperature at free surface, $\Sigma = \Sigma_0 - \gamma_T\beta d$, where γ_T is commonly a positive constant. In addition, the Biot number, $Bi = hd/\epsilon$ arises from the heat transfer (cooling) at the upper boundary according to Newton's law, with $\epsilon = \kappa c_H$ the heat conductivity of the liquid and h the heat transfer coefficient. For a perfectly heat conducting surface Bi tends to infinity, while for an adiabatically insulating boundary it goes to zero.

On the other hand, in the case of a finite magnetic permeability χ_b of the boundaries, the scalar magnetic potential must satisfy the following dimensionless boundary conditions[5]

$$(1+\chi_b)(D\,\Phi - \Theta) \pm k\,\Phi = 0, \quad (18)$$

at $z = \pm 1/2$. The occurrence of temperature variations in the magnetic boundary condition at the free boundary (at $z = -1/2$ there is $\Theta = 0$) is specific for the combination of surface and magnetic effects. The effective surface susceptibility $\chi_b = \chi - (1+\chi)(M_3 - 1)$ is slightly smaller than the linear one. Note that in the limit when χ_b tends to infinity, Eqs. (18) gives $D\Phi = \Theta$, which is often used as a simplified boundary condition.

The Marangoni instability is a capillary surface effect and driven by the applied temperature difference that shows up in Ma, in contrast to the Bénard instability that is a bulk instability driven by the applied temperature difference that shows up in Ra. Since Ra and Ma both depend on the applied temperature difference, it is better to define two new parameters, η (*rate of heating*) and α (*gravity parameter*), defined via $Ra = \eta\alpha Ra_0$ and $Ma = (1-\alpha)\eta Ma_0$. Here, $Ra_0 = 669$

and $Ma_0 = 79.607$ are the critical Rayleigh number obtained in the absence of capillarity for Newtonian fluids and the critical Marangoni number obtained in the absence of gravity for Newtonian fluids with an adiabatically isolated upper surface, respectively. The rate of heating η is proportional to the applied temperature gradient and acts as control parameter, while α measures the ratio between the gravity and the capillarity effect[12], as can be seen from the inverse relations

$$\eta = \frac{Ra}{Ra_0} + \frac{Ma}{Ma_0} \tag{19}$$

and

$$\alpha = \frac{RaMa_0}{MaRa_0 + RaMa_0}. \tag{20}$$

In particular, the case $\alpha = 0$ corresponds to a pure capillary effect ($Ra = 0$), while the case $\alpha = 1$ describes a pure gravity effect ($Ma = 0$). In the following, we will calculate, for some fixed values of α, the critical value η_c of η, where the instability sets in.

In order to solve Eqs. (13) - (15) with these realistic boundary conditions, we use a spectral collocation method. Spectral methods ensure an exponential convergence to the solution and are the best available numerical techniques for solving simple eigenvalue – eigenfunction problems. Here, we follow the technique of collocation points on a Chebyschev grid as described by Threfethen.[20] The collocation points (Gauss–Lobato) are located at height $z_j = \cos(j\pi/N)$ where the index j runs from $j = 0$ to $j = N$. Note that here the z-variable ranges from -1 to $+1$ and one has therefore to rescale Eqs. (13) - (15) accordingly, because the physical domain is defined in the range $[-1/2, +1/2]$. We use $N = 20$ collocation points in the vertical direction, for which the equations and the boundary conditions are expressed. More collocation points only modify the sixth significant digit of the result. By using the collocation method, the set of differential equations (13) - (15) is transformed into a set of linear algebraic equations. The eigenfunctions $\{\Theta(z), \Phi(z), W(z)\}$ are transformed into eigenvectors defined at the collocation points. After this stage of discretization, one is left with a classical generalized eigenvalue–eigenvector problem.

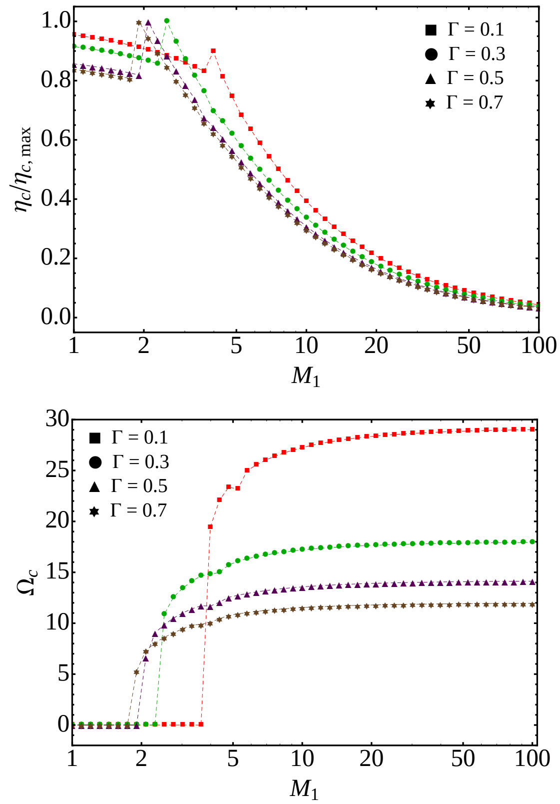

Figure 1: η_c (top) and Ω_c (bottom) as a function of M_1 for different values of Γ at $\alpha = 0.25$. The different curves belong to different Deborah numbers (red squares = 0.1, green dots = 0.3, purple triangles = 0.5, brown stars = 0.7). The other fixed parameters are $Pr = 10$, $\Lambda = 0.5$, $M_3 = 1.1$, $Bi = 10^{-6}$, and $\chi_b = 1$.

In the case of the oscillatory instability considered here, choosing a fixed value of the horizontal wavenumber k, one looks for a marginally stable solution of η (with $\Sigma = 0$) making sure that η and Ω are real quantities. This procedure is repeated for several values of k leading to the marginal stability curve η versus k. The minimum of this curve gives η_c and k_c, and the corresponding value of Ω is the critical fre-

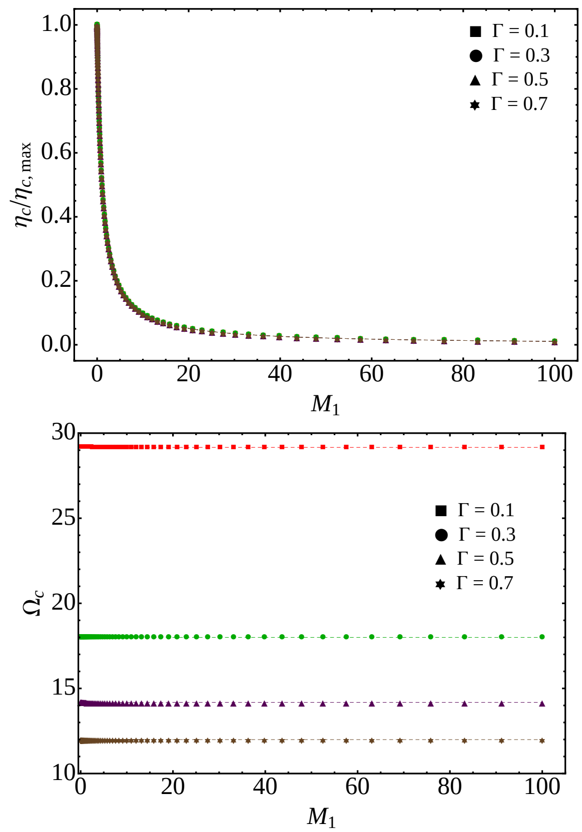

Figure 2: η_c (top) and Ω_c (bottom) as a function of M_1 for different values of Γ at $\alpha = 1$. The other fixed parameters are the same of Figure 1.

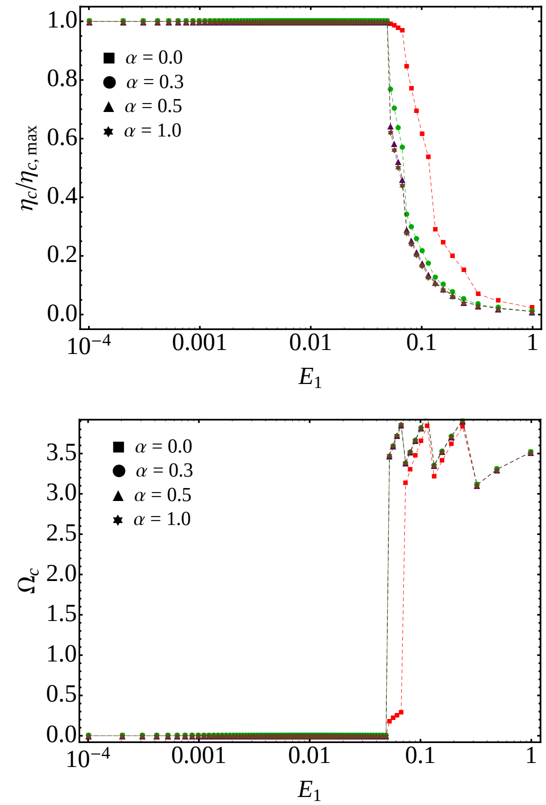

Figure 3: η_c (top) and Ω_c (bottom) as a function of E_1 for different values of α (red squares = 0, green dots = 0.3, purple triangles = 0.5, brown stars = 1) . The fixed parameters are $Pr = 10$, $\Gamma = 100$, $M_1 = 10$, $M_3 = 1.1$, $Bi = 10^{-6}$, and $\chi_b = 1$.

quency Ω_c.

The main results are given in Figs. 1 to 3. In all cases the critical heating rate, η_c, and its corresponding frequency, Ω_c, are displayed as a function of different sets of control and material parameters. In particular, we concentrate the discussion on the influence of the magnetic and viscoelastic properties on those quantities. Figure 1 and 2 show η_c and Ω_c as a function of the magnetic number $M_1 \sim H_0^2$ for four different Deborah numbers. In Fig. 1 the Marangoni aspect is manifest ($\alpha = 0.25$), while in Fig. 2 the pure Bénard case ($\alpha = 1$) is shown for comparison. In both cases we find that the magnetic field destabilizes the system, since the critical value of the threshold, η_c, decreases when M_1 increases. In the first case, with $\alpha = 0.25$, the decrease is a rather moderate when compared to the the pure Bénard case ($\alpha = 1$). In both cases, however, the threshold is independent of the Deborah number Γ. On the other hand, the critical frequency is a non-monotonous function of the magnetic field with a shallow minimum at a certain value of M_1 for the Marangoni case (Fig. 1). This feature vanishes completely for $\alpha \to 1$, where Ω_c is constant with respect to M_1, as is shown in Fig. 2. The constant value of Ω_c, however, depends strongly on the Deborah number for both cases, and decreases for increasing Γ. This simply reflects the expectation that a slower relaxation of the elasticity enhances the internal time scale for the critical perturbations. (Note that for fixed Λ an increasing Γ means a decreasing E_1).

Finally, we remark that, for this range of parameters, the influence of the other magnetic number, M_3 is irrelevant, because η_c and Ω_c change less than 0.05%.

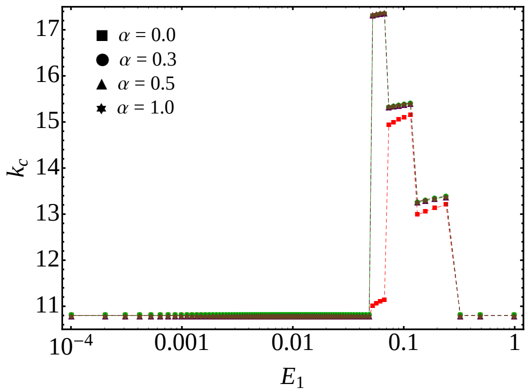

Figure 4: The critical wavenumber k_c as a function of E_1 for different values of α (as in Fig. 3). The fixed parameters are as in Fig. 3

Figure 3 shows η_c and Ω_c as a function of the elastic modulus E_1 for different values of the gravity parameter α. Obviously, when increasing the elastic modulus, there is a (hard mode) transition from the stationary instability to an oscillatory one, with a finite critical frequency at the transition. Such a transition has to be expected, since for a Newtonian fluid ($E_1 \rightarrow 0$) only the stationary instability is possible, while in the elastic case (E_1 large) convection cannot be stationary, but only oscillatory. This transition occurs at a smaller elastic modulus in the Marangoni case than in the Bénard one. In addition, we observe that in the oscillatory regime the frequency is a non-smooth function of Λ, a known phenomenon in viscoelastic fluids with complex boundary conditions.[12,21,22] It is directly related to jumps in the critical wavenumber k_c (Fig. 4). At the transition the horizontal width of the spatial patterns (e.g. convection rolls) decreases and reaches again the stationary value in a few steps when increasing E_1. Above that, Ω_c is a smooth increasing function of E_1 as one expects from an almost elastic medium. (The time scale of the oscillations is much smaller than the relaxation time of the elasticity). Finally, let us comment that this phase transition is robust against changes of the Deborah number within a wide range. The critical heating rate is independent of the elastic properties for the stationary instability, but decreases in the oscillatory one rather rapidly with increasing elasticity.

CONCLUSIONS

In the present work, Bénard-Marangoni convection in a magnetic viscoelastic liquid is studied. The stability thresholds have numerically been determined by the spectral method. The technique of collocation points (Gauss−Lobato) as described by Threfethen[20] was used. Due to the presence of various destabilizing effects, i.e. buoyancy and magnetic forces, and of additional relaxation channels due to the linear viscoelasticity, the discussion of the stability curves becomes rather intricate. We found that a magnetic field destabilizes the system in the Marangoni case less than in the Bénard one, while the critical frequency depends strongly on the Deborah number in both cases. The critical thresholds are independent of the Deborah number. As a function of the elastic modulus the oscillatory instability, whose critical frequency is non-smooth, is competing with the stationary one. Let us finally comment that, very often ferrofluids show a finite separation ratio and a finite magnetic separation ratio and, therefore, require a binary mixture description. However, for materials, where the separation ratio and magnetic separation ratio are not too large, the simple fluid approximation is valid[6]. The present work is based on this last approximation. A detailed study on the oscillatory bifurcation for magnetic binary mixtures is still in progress.

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