

Soret-driven thermal instability  
in ferrofluid binary mixtures

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<http://www.mpi-p-mainz.mpg.de/~pleiner/instab.html>

ferrofluids/-gels: ..... /ferro.html

rheology: ..... /rheo.html

liquid crystals: ..... /banana.html

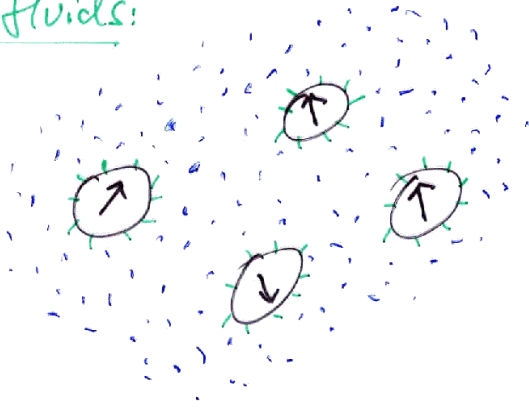
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1) Introduction - ferrofluids  
- binary mixtures

2) Thermal instability without magnetic field  
- threshold  
- linear growth rate  
- nonlinear amplitude

3) Thermal instability with magnetic field  
- basic equations  
- numerical solutions  
- analytical  
- boundary layers

Ferrofluids:



magnetic particles (10 nm) in carrier fluid  
 sterically or charge stabilized  
 superparamagnetic: high  $M_s$  for low  $H_{ext}$   
 black

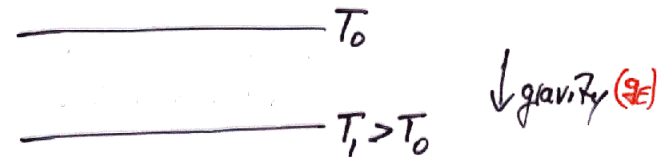
applications: seal in hard drives  
 active clumping  
 cancer therapy

science: new types of instabilities / pattern formation

developments: ferro-nematics  
 ferro-gels

thermal instability:

single fluid:



$$Ra \sim \frac{\alpha_0 \beta g E}{\kappa \nu}$$

$\alpha_0$  thermal expans  
 $\beta$  temp gradient  
 $\kappa$  heat conduct  
 $\nu$  viscosity

$Ra \geq Ra_c$  convection pattern formation

binary fluid: 2 conserved masses, 1 velocity

concentration current  $\vec{j} = D \vec{\nabla} c + D_T \vec{\nabla} T$

diffusion      thermal diffusion (Soret)

solvent expansion  $\frac{\delta \rho_c}{\rho_0} = \alpha_c \delta c - \alpha_\theta \delta T$

Lewis number  $L \sim \frac{D}{\kappa} \ll 1$

separation ratio  $\psi \sim \frac{\alpha_c}{\alpha_\theta} \frac{D_T}{D} \gg 1$

II) Thermal instability - no field

$$\operatorname{div} \vec{v} = 0$$

$$\partial_t \vec{v} + (\vec{v} \cdot \nabla) \vec{v} = -\vec{\nabla} p + Pr \Delta \vec{v} + Pr Ra [\delta T - \psi \delta c] \vec{e}_z$$

$$\partial_t T + (\vec{v} \cdot \nabla) T = \Delta T$$

$$\partial_t c + (\vec{v} \cdot \nabla) c = L(\Delta c + \Delta T)$$

$$\vec{v}|_b = 0$$

$$T|_b = \bar{T} + \frac{1}{2}$$

$$\nabla_z c + \nabla_z T|_b = 0$$

pure conduction, no convection:

$$\vec{v} = 0$$

$$T_{\text{cond}} = \bar{T} - z$$

$$c_{\text{cond}} = \bar{c} + z$$

however: this state is never reached!

$$L \ll 1!$$

instead:  $C = C_0$  good for  $t \ll L^{-1}$   
outside boundary layer

linear stability analysis, growth rate  $\lambda = 0$

$$\hookrightarrow Ra_c = \frac{1}{1+\psi} Ra_c^0$$

$$\ll Ra_c^0$$

however: instability unobservable near  $Ra_c$   
 $\lambda(Ra) \ll L^{-1}$  required!  
 $\lambda > L$

$\Rightarrow$  calculate linear growth rate  $\lambda(Ra)$   
calculate nonlinear (velocity) saturation amplitude

linear growth rate:

deviations:  $W = A(z) \cos kz + \cos^2 \pi z$

$\theta = B(z) \cos kz + \cos^2 \pi z$

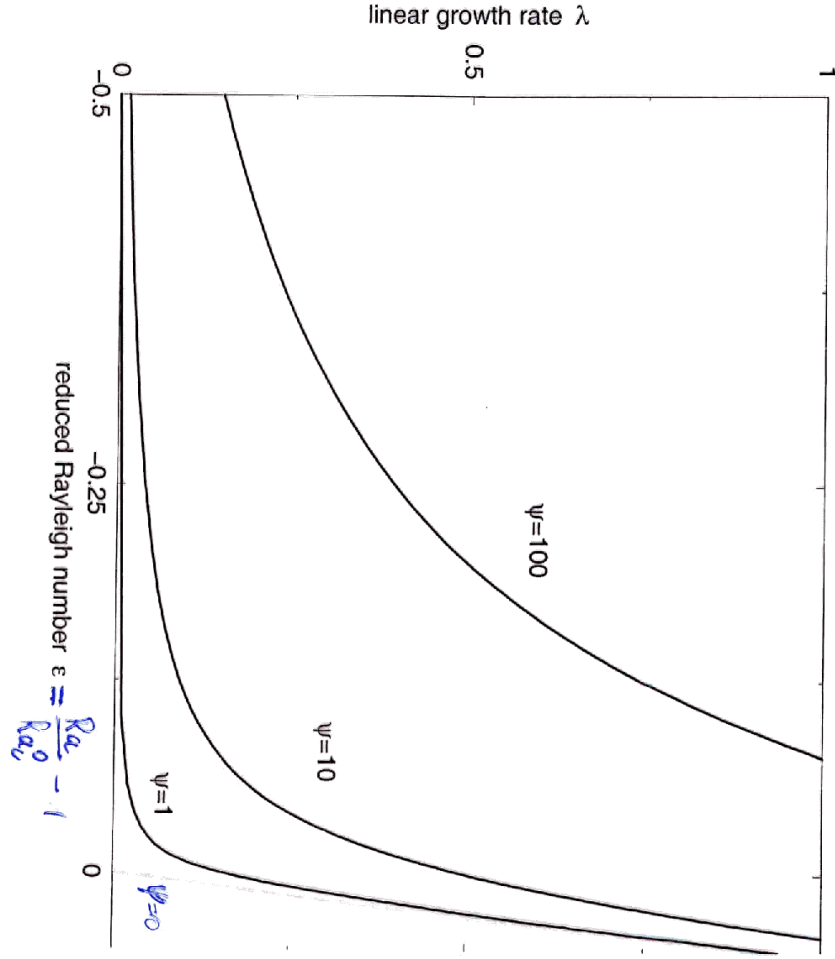
$C = -\theta + \cos kz + \sum_{n=2}^{\infty} b_n(z) \cos 2n\pi z$

boundary conditions, symmetric  $\phi_h$ .

Galerkin

approximate analytical solution ( $\lambda \gg 1, \psi \gg 1$ )

$$3 Ra Pr (\lambda + 2\pi^2 \psi) = (\lambda + 2\pi^2)(27\pi^2 Pr + 7\lambda) \quad (k = \pi)$$



saturation amplitude:

$$C(x, z, t) = C_0(z, t) + c_1(z, t) \cos kx$$

$$T(x, z, t) = \theta_0(z, t) + \theta_1(z, t) \cos kx$$

$$v_z(x, z, t) = w_1(z, t) \cos kx$$

with  $w_1(z, t) = A(t) \cos^2 \pi z$

$$\theta_1(z, t) = B(t) \cos \pi z$$

$$\theta_0(z, t) = f(t) \sin 2\pi z$$

$$C_0(z, t) = z - \theta_0(z, t) + \sum_{n=0}^N a_n(t) \sin(2n+1)\pi z$$

$$c_1(z, t) = -\theta_1(z, t) + \sum_{n=0}^N b_n(t) \cos 2n\pi z$$

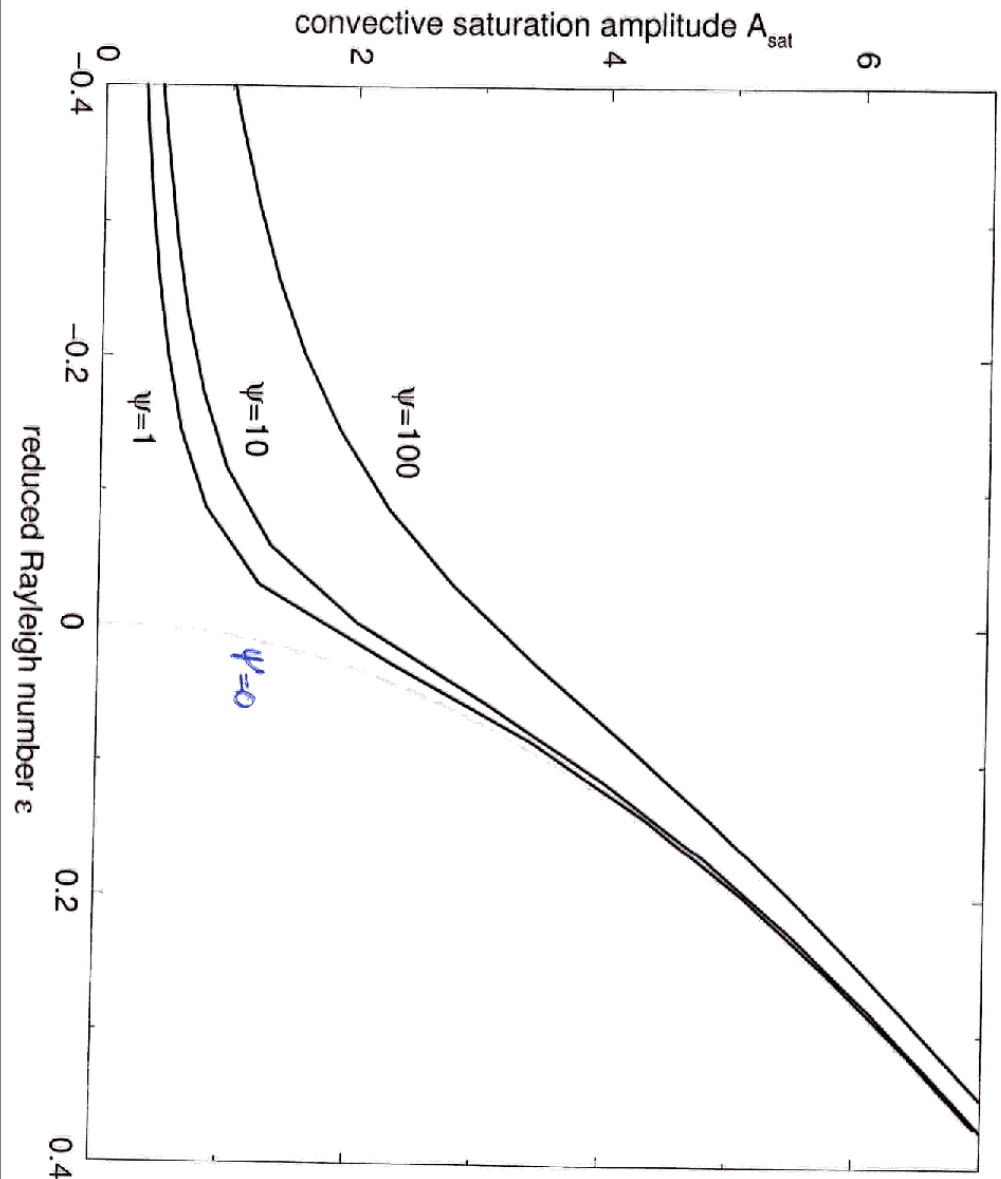
b.c. ok

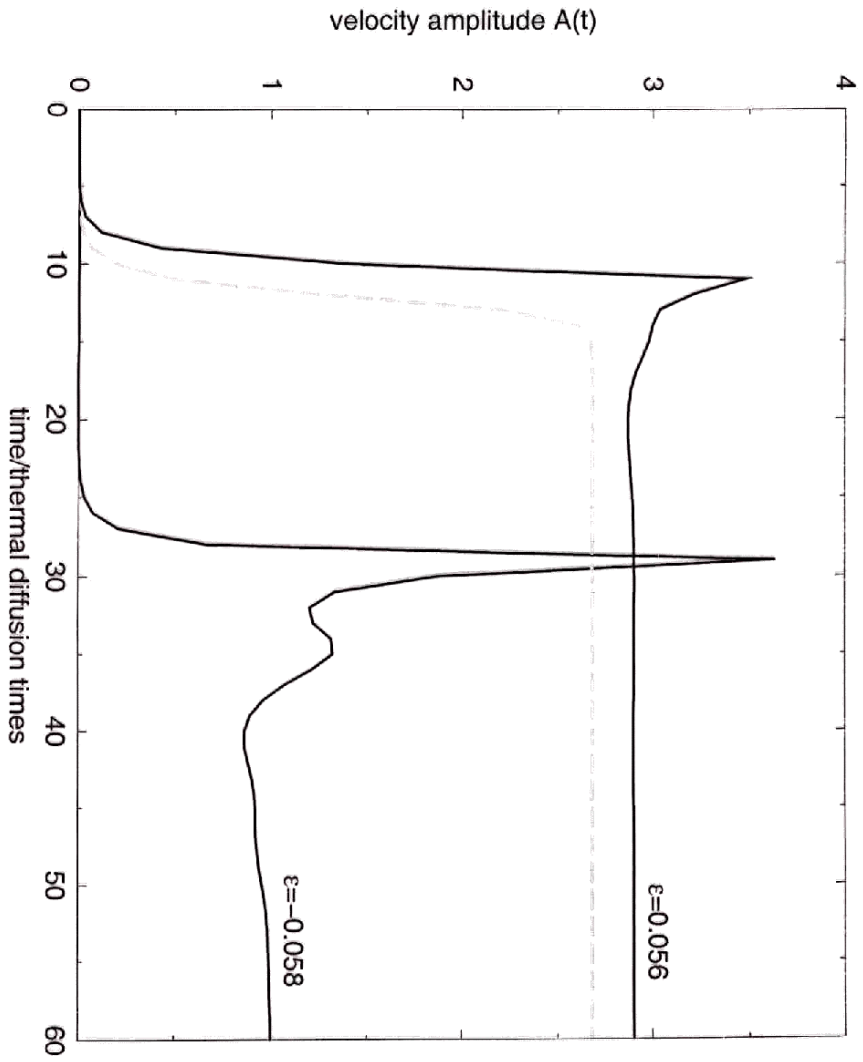
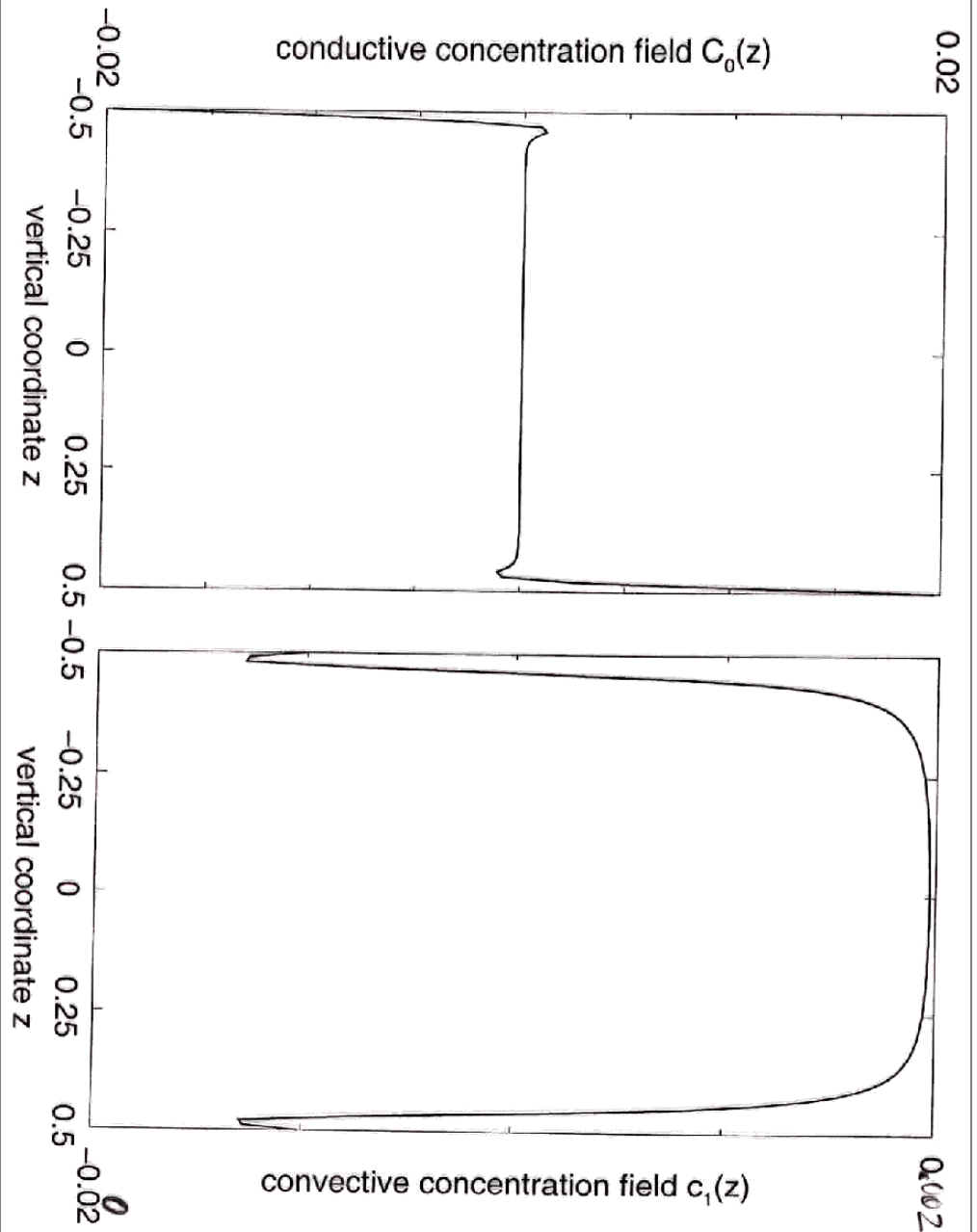
for  $\psi=0$   $\rightarrow$  3 mode Lorenz model

$N \approx 20$  (concentration boundary layer)

$k = 3 \dots 3.5$  ( $\pi!$ )

no oscillatory inst. found





Summary part I: there is a forward bifurcation into rolls also for  $Ra \lesssim Ra_c^0$ , but unobservable for  $Ra_c \approx Ra \ll Ra_c^0$  (exp. S. Odenthal [Bremen])

II. Thermal instability with magnetic field:

field effects: Maxwell eq.  $\text{div } \vec{B} = 0, \text{curl } \vec{H} = 0, + \text{b.c.}$  ✓

Kelvin force  $M_j \nabla H_j$  in Navier Stokes ✓

magnetoforesis  $\delta \vec{B} = \bar{\epsilon}(\vec{H}) \delta \vec{H} + H_0 [\chi_T \delta T + \chi_c \delta c]$

$$\begin{aligned} \delta T &= \dots + \chi_T \vec{H}_0 \cdot \delta \vec{H} \\ \delta \mu_c &= \dots + \chi_c \vec{H}_0 \cdot \delta \vec{H} \end{aligned} \quad (\checkmark)$$

volume expansion  $\partial \rho / \rho = \dots + \alpha_H \vec{H} \cdot \delta \vec{H}$  —

Hall-like effects:  $D_{ij} = D_0 \delta_{ij} + D^R \epsilon_{ijk} H_k$  —

ground state:

$$\begin{aligned} \vec{v} &= 0 \\ T &= T_0 - \beta z \\ C &= C_0 \\ H_z &= H_0 \left( 1 + \frac{\chi_T}{\bar{\epsilon}} \beta z \right) \end{aligned}$$

← flat concentration profile

deviations:  $\vec{v}, \theta, c, \vec{H} = H_z \hat{e}_z - \nabla \phi, \vec{H}_{\text{ext}} = H_0 - \nabla \psi$

Simple rolls  $\sim \cos kx$ ,  $M_4 = M_5 = F = 0$

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Simple Galerkin solution

sorting for different lateral dependencies yields the following system of equations

$$\frac{1}{Pr} \partial_t (\nabla_z^2 - k^2) w_1 = -Ra k^2 [(1 + M_1)\theta_1 - (\psi + M_1\psi_m)c_1 - M_1\nabla_z\phi_1] + Ra M_1 k^2 (\theta_1 - \psi_m c_1 - \nabla_z\phi_1) \nabla_z (\theta_0 - \psi_m c_0) + (\nabla_z^2 - k^2)^2 w_1, \quad (3.58)$$

$$\partial_t c_0 + \frac{1}{2} \nabla_z (w_1 c_1) = L \nabla_z^2 [(1 + M_2\psi_m)c_0 + (1 - M_2)\theta_0], \quad (3.59)$$

$$\partial_t c_1 + w_1 \nabla_z c_0 = L (\nabla_z^2 - k^2) (c_1 + \theta_1 - M_2 \nabla_z \phi_1), \quad (3.60)$$

$$\partial_t \theta_0 + \frac{1}{2} \nabla_z (w_1 \theta_1) = \nabla_z^2 \theta_0, \quad (3.61)$$

$$\partial_t \theta_1 + w_1 \nabla_z \theta_0 = -w_1 + (\nabla_z^2 - k^2) \theta_1, \quad (3.62)$$

$$(\nabla_z^2 - M_3 k^2) \phi_1 = \nabla_z (\theta_1 - \psi_m c_1), \quad (3.63)$$

The field  $\phi_0$  has already been eliminated with the help of  $\nabla_z^2 \phi_0 = \nabla_z (\theta_0 - \psi_m c_0)$ . This has also been used to write the remaining boundary conditions as

$$\nabla_z (c_1 + \theta_1 - M_2 \nabla_z \phi_1)|_{z=\pm 1/2} = 0, \quad (3.64)$$

$$\nabla_z ((1 + M_2\psi_m)c_0 + (1 - M_2)\theta_0)|_{z=\pm 1/2} = 1 - M_2, \quad (3.65)$$

$$\theta_1|_{z=\pm 1/2} = \theta_0|_{z=\pm 1/2} = 0, \quad (3.66)$$

$$w_1|_{z=\pm 1/2} = \nabla_z w_1|_{z=\pm 1/2} = 0, \quad (3.67)$$

To solve this boundary-value problem we adopt vertical profiles  $w_1, \theta_0, \theta_1, c_0, c_1$  and  $\phi_1$  in the form

$$w_1(z, t) = A(t) \cos^2(\pi z), \quad (3.68)$$

$$\theta_1(z, t) = B(t) \cos \pi z, \quad (3.69)$$

$$\theta_0(z, t) = G(t) \sin 2\pi z, \quad (3.70)$$

multi-mode boundary layer

$$c_0(z, t) = \frac{1 - M_2}{1 + \psi_m M_2} (z - \theta_0(z, t)) + \sum_{n=0}^{n=N} a_n(t) \sin(2n + 1)\pi z, \quad (3.71)$$

$$c_1(z, t) = -\theta_1(z, t) + \sum_{n=0}^{n=N} b_n(t) \cos 2n\pi z, \quad (3.72)$$

$$\phi_1(z, t) = A_0(t) z + \sum_{n=0}^{n=N_1} \frac{A_n(t) \sin 2\pi n z}{2\pi n} \quad (3.73)$$

which satisfy the boundary conditions (3.56, 3.64-3.67) identically, if  $A_0(2+k) + \sum_{n=1}^{N_1} (-)^n A_n + \psi_m \sum_{n=1}^N (-)^n b_n = 0$  is chosen.

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Simple Galerkin solution

temperature from Eq. (3.36) is  $\theta$ . Then Eqs. (3.5), and (3.22)-(3.25) lead to

$$\nabla \cdot \mathbf{v} = 0 \quad (3.39)$$

$$\left[ \frac{\partial}{\partial t} + (\mathbf{v} \cdot \nabla) \right] (\theta - M_4 \nabla_z \phi) = w(1 - M_4) + \Delta \theta + F \Delta (C - M_2 \nabla_z \phi) \quad (3.40)$$

$$\left[ \frac{\partial}{\partial t} + (\mathbf{v} \cdot \nabla) \right] C = L \Delta (\theta + C - M_2 \nabla_z \phi) \quad (3.41)$$

$$\frac{1}{Pr} \left[ \frac{\partial}{\partial t} + (\mathbf{v} \cdot \nabla) \right] (\text{curl} \mathbf{v})_i = Ra \epsilon_{ikz} \nabla_k [(1 + M_1)\theta - (\psi + \psi_m M_1)C + (M_5 - M_1)\nabla_z \phi] - Ra M_1 \epsilon_{ikl} (\nabla_l \nabla_z \phi) \nabla_k (\theta - \psi_m C) + \Delta (\text{curl} \mathbf{v})_i \quad (3.42)$$

$$(\nabla_z^2 + M_3 \Delta_{\perp}) \phi = \nabla_z (\theta - \psi_m C) \quad (3.43)$$

$$\Delta \phi_e = 0 \quad (3.44)$$

where  $w$  is the  $z$  component of the velocity. The transverse Laplacean  $\Delta_{\perp} = \Delta - \nabla_z^2$ . The non-dimensional parameters introduced here are: the Rayleigh number  $Ra = \alpha_0 \beta_0 g_E d^4 / (\kappa \nu)$ , the Prandtl number  $Pr = \nu / \kappa$ , the separation ratio  $\psi = \alpha_c \rho_0 \bar{D}_T / (\alpha_0 \gamma_H D)$ , the magnetic separation ratio  $\psi_m = -\chi_c \rho_0 \bar{D}_T / (\chi_T \gamma_H D)$ , the Lewis number  $L = \gamma_H c_H D / (\rho_0^2 \bar{\kappa} T_0) = D_c / \kappa$ , the strength of magnetic force relative to buoyancy  $M_1 = \beta_0 \chi_T^2 H_0^2 / (\rho_0 g_E \alpha_0 \bar{\epsilon})$ , the magnetophoretic number  $M_2 = D \chi_c \chi_T H_0^2 / (\rho_0 \bar{D}_T \bar{\epsilon})$ , the nonlinearity of magnetization  $M_3 = (1 + \chi_0) / \bar{\epsilon} \approx 1 - \chi_H H_0^2 / (1 + \chi_0)$ , the relative strength of the temperature dependence of the magnetic susceptibility  $M_4 = \chi_T^2 H_0^2 T_0 / (c_H \bar{\epsilon})$ , the ratio of magnetic to thermal buoyancy  $M_5 = \alpha_H \chi_T H_0^2 / (\alpha_0 \bar{\epsilon})$ , and the Dufour number  $F = \bar{D}_T^2 / (D \bar{\kappa}) = D_s D_f / (\kappa D_c)$ . The stability conditions (3.20) require  $M_4 < 1$  and  $M_2 \psi_m > -1$ .

According to our choice of 'rigid' and ideally conducting boundaries, the boundary conditions for the deviations from the conducting state read

$$\theta|_{z=\pm 1/2} = 0 \quad (3.45)$$

$$w|_{z=\pm 1/2} = 0 \quad (3.46)$$

$$\nabla_z w|_{z=\pm 1/2} = 0 \quad (3.47)$$

$$\nabla_z (\theta + C - M_2 \nabla_z \phi)|_{z=\pm 1/2} = 1 - M_2 \quad (3.48)$$

and the magnetic boundary conditions (3.28), (3.29) are

$$\bar{\epsilon} (\nabla_z \phi + \psi_m C) - \nabla_z \phi_e|_{z=\pm 1/2} = 0 \quad (3.49)$$

$$\nabla_{\perp} \phi - \nabla_{\perp} \phi_e|_{z=\pm 1/2} = 0 \quad (3.50)$$

These boundary conditions close the problem to find the fields  $\mathbf{v}, \theta, C, \phi$  and  $\phi_e$ .

5 M's ~ H\_0^2

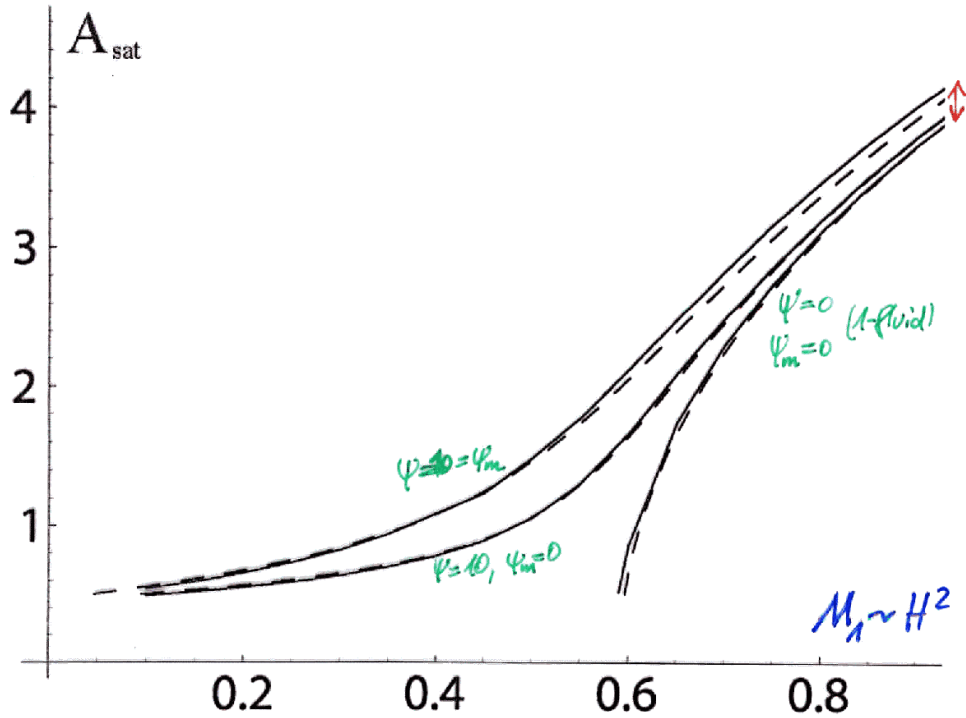
b.c. couple C with phi



$N=50$

$M_2=0$  (Kelvin force only)

$Ra \approx 1300 \leq Ra_c^0$



approximate analytical solutions

boundary layer problem for concentration and magnetic potential

boundary layer depth  $\sim L^{1/3}$

$$\frac{18\pi^4}{Ra} = \frac{1 + M_1(\beta - 2\pi\bar{\beta}\zeta(A^2))}{1 + \frac{3}{40\pi^2}A^2}$$

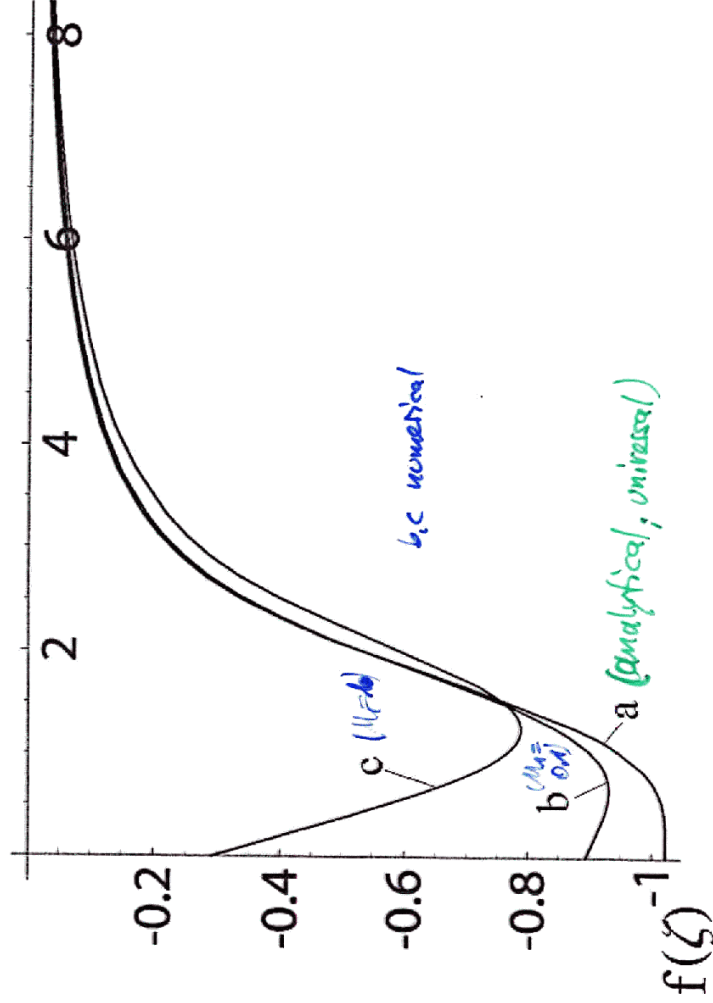
$$+ (1-M_2)\frac{32\pi^2}{3A^2}\left[L(\psi + M_1\psi_m) + \gamma(1-\bar{\beta}\zeta(A^2))M_1\psi_m\left(\frac{L^2A}{1+\psi_m M_2}\right)^{1/3}\right]$$

with  $\zeta(A^2) = \frac{9A^2}{160\pi^3(1 + \frac{3}{40\pi^2}A^2)}$

magnetoforesis:  $M_2 < 0$ ;  $M_2 \geq \frac{-1}{\psi_m}$  thermodynamic stability condition

(strong) influence only for  $M_2\psi_m \rightarrow -1$   
or  $H^2 \rightarrow \beta_0 \bar{\epsilon} \bar{\chi}_c^{-1}$

boundary layers ( $M_2=0, \psi = \psi_m = 10$ )



Conclusion II: External field makes the "imperfection" of the bifurcation more pronounced

pronounced boundary layer profiles that couple to the bulk behavior

for varying fields breakdown of 'binary mixture' description: agglomeration?

effectively  $\partial_z c = L(1 + \chi_m \mu) \nabla^2 c$

future: negative  $\psi$ ; oscillatory instability? patterns other than rolls?