Fluid-fluid phase transitions in ER-plasmas: Sheets & Strings

P. Brandt, A. V. Ivlev and G. E. Morfill

Max-Planck-Institut für extraterrestrische Physik
Gruppe Theorie & komplexe Plasmen

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Tunable microscopic structures

- buzzword: *smart materials*: materials with properties that can be changed in a controlled fashion
  - well known: liquid crystals (tunable property: isotropic, nematic, smectic, . . .)
  - electro- and magnetorheological systems (ER/MR)
    tunable property: **structure**

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- found at various scales
  colloids, complex plasmas, nano-sized particles (proteins, polymers), mesoscopic level, . . .

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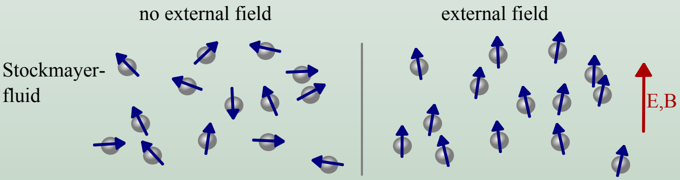
- found at various scales
  colloids, complex plasmas, nano-sized particles (proteins, polymers), mesoscopic level, . . .
- a toy system for critical phenomena

Electro- and magnetorheological Systems (ERMR)

A short retrospection:
permanent dipoles (embedded in media) (also called: Stockmayer-fluids)
Interaction:

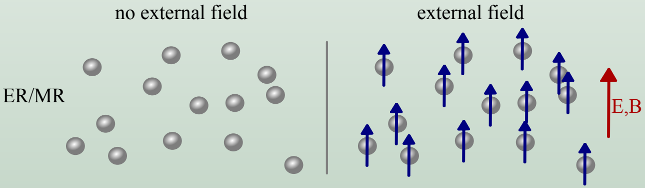
$$u_{dipol}(i,j) = -\frac{(\mu_i \cdot \mathbf{r})(\mu_j \cdot \mathbf{r}) - (\mu_i \cdot \mu_j) r^2}{r^5}$$



Electro- and magnetorheological Systems (ERMR)

Electro- or magnetorheological fluids: particles with *induced* moments (may be both magneto or electrostatic) in media:
Interaction:

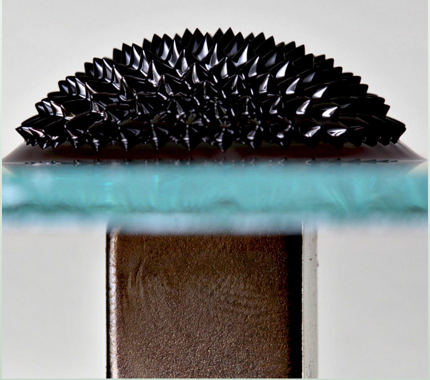
$$u_{rheo}(i,j) = -m^2 \frac{(\mathbf{e}_z \cdot \mathbf{e}_r)^2 - 1}{r^3} = -\xi \frac{1}{r^3} P_2(cos\theta)$$



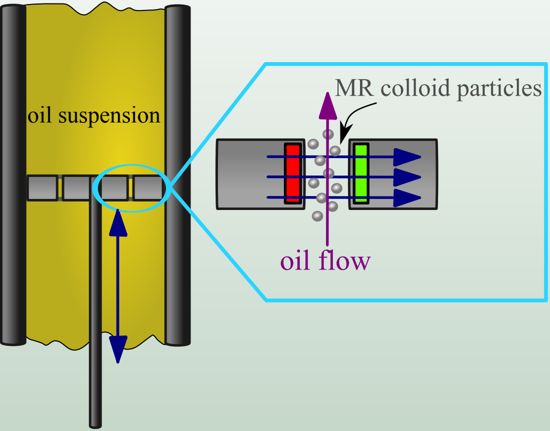
ER/MR

(MRF fluid video)

MR fluid:



ER/MR | Applications



smart shock absorbers

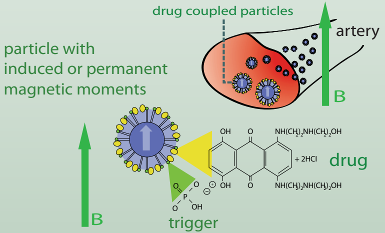
i.e. used in production model car Audi *R8*
"audi magnetic ride"

picture: wikimedia.org

ER/MR | Applications

Other concepts...

- smart shock absorbers R. Stanway et al., Smart Materials 5 (1996) 464
- a tool for biological chemistry and medicine **magnetofection** C. Plank et al., Bio. Chem. 384 (2003) 737
- electrostatic switchable **valves** S.B. Choi et al., Smart Materials 14 (2005) 1483
- tunable **photonic crystals** R. Tao et al., APL 93 (2008) 241105



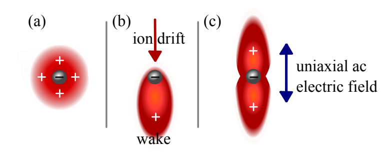
Bad news...

German Federal Environmental Agency **warns** of nano products...

October 21^{th} 2009

ER plasmas

Designable potentials



generic potential:

$V(\mathbf{r}) = \epsilon\left[\phi_{\mathrm{I}}(r) - \xi\phi_{\mathrm{A}}(r)P_2(\cos\theta)\right]$

basic principle:
application of rf discharge

$\longrightarrow$ tunable "dipolar" interaction:

$$W(r,\theta) = Q^2\left[\frac{e^{-r/\lambda}}{r} - \frac{M_{\mathrm{T}}^2\lambda^2}{r^3}P_2(\cos\theta)\right]$$

(other designed potentials also possible)[1]

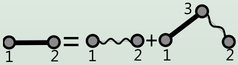
[1] R. Kompaneets, PoP 14(2007)052108

Theoretical framework

Ornstein-Zernike equation (OZ)

defines a basic relation for *direct* $c(1,2)$ and *total* h(1,2) correlation function
(total correlation function: $h(\mathbf{r}) = g(\mathbf{r}) - 1$)

$$h(1,2) = c(1,2) + \rho \int d(3) c(1,3) h(3,2)$$



$\rightarrow$ ☐ open integral equation
$\rightarrow$ ☐ closure relation required

$$g(1,2) = \exp\left(-\beta\phi(1,2) + h(1,2) - c(1,2) + B(1,2)\right)$$

appropriate choice $B(1,2) = 0$
(hypernetted chain closure)

Theoretical framework

Basic solution (isotropic fluids)

- isotropic systems: $g(\mathbf{r}) = g(|r|)$
- $h(r) = c(r) + \rho \int dr' c(r') h(|r - r'|)$

Theoretical framework

Basic solution (isotropic fluids)

- isotropic systems: $g(\mathbf{r}) = g(|r|)$
- $h(r) = c(r) + \rho \int dr' c(r') h(|r - r'|)$
- Fourier transformed eq. $$\underbrace{(\hat{h} - \hat{c})(k)}_{\hat{\gamma}(k)} = 4\pi\rho\hat{h}(k)\hat{c}(k)$$
- *plus* closure relation $c(r) = F[r, \gamma(r), V(r)]$
- usually solved using iterative approaches (i.e. Picard iterations)

OZ | Anisotropic systems?

- **Stockmayer-fluids:** Blum[2], Fries[3]:
  Solution of OZ for anisotropic binary interactions but rotational invariant systems
- **ERMR** (Legendre decomposition):

$$c(\mathbf{r}) = \sum_l c_l(r) P_l(\cos\theta) \qquad h(\mathbf{r}) = \sum_l h_l(r) P_l(\cos\theta)$$

Hankel-Fourier-Transformation...

$$\hat{h}(\mathbf{k}) - \hat{c}(\mathbf{k}) = (4\pi)^2 \rho \sum_n d_n(k) P_k(\cos\alpha)$$

$$d_n(k) = (2n+1) \sum_{l,l'} \begin{pmatrix} l & l' & n \\ 0 & 0 & 0 \end{pmatrix}^2 i^{l+l'} \hat{h}_l(k) \hat{c}_{l'}(k).$$

[2] Blum et al., JCP 56 (1972) 303
[3] Fries and Patey, JCP 82 (1985) 429

OZ | Anisotropic systems?

- skipping all terms except ($l = 0, 2, 4$)
- OZ kernel (now a *linear* equation)

$$\frac{\hat{h}_0 - \hat{c}_0}{4\pi\rho} = \hat{h}_0\hat{c}_0 + \frac{1}{5}\hat{h}_2\hat{c}_2 + \frac{1}{9}\hat{h}_4\hat{c}_4,$$

$$\frac{\hat{h}_2 - \hat{c}_2}{4\pi\rho} = \hat{h}_2\hat{c}_0 + \hat{h}_0\hat{c}_2 - \frac{2}{7}\hat{h}_2\hat{c}_2 + \frac{2}{7}(\hat{h}_4\hat{c}_2 + \hat{h}_2\hat{c}_4) - \frac{100}{693}\hat{h}_4\hat{c}_4,$$

$$\frac{\hat{h}_4 - \hat{c}_4}{4\pi\rho} = \hat{h}_4\hat{c}_0 + \hat{h}_0\hat{c}_4 + \frac{18}{35}\hat{h}_2\hat{c}_2 - \frac{20}{77}(\hat{h}_4\hat{c}_2 + \hat{h}_2\hat{c}_4) + \frac{162}{1001}\hat{h}_4\hat{c}_4,$$

- isotropic case: $\hat{c}_2 = 0,\ \hat{c}_4 = 0$
- three component extension

OZ | Closure relation

- remember("closure relation") :

$$g(\mathbf{r}) = \exp\left[-\beta V(\mathbf{r}) + h(\mathbf{r}) - c(\mathbf{r}) + B(\mathbf{r})\right] \qquad (\text{HNC: } B \equiv 0)$$

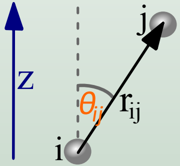
- anisotropic system:

$$g(\mathbf{r}) = \sum_{l=0,2,4} g_l(r) P_l(\cos\theta)$$
$$= \exp\Big(\sum_l (-\beta\phi_l(r) + \gamma_l(r)) P_l(\cos\theta)\Big)$$

- Solution: [OZ Kernel, Closure (HNC)] → iterative solver (Gillan-Labik) → $g(\mathbf{r}) = \sum_l g_l(r) P_l(\cos\theta)$

Monte Carlo simulation

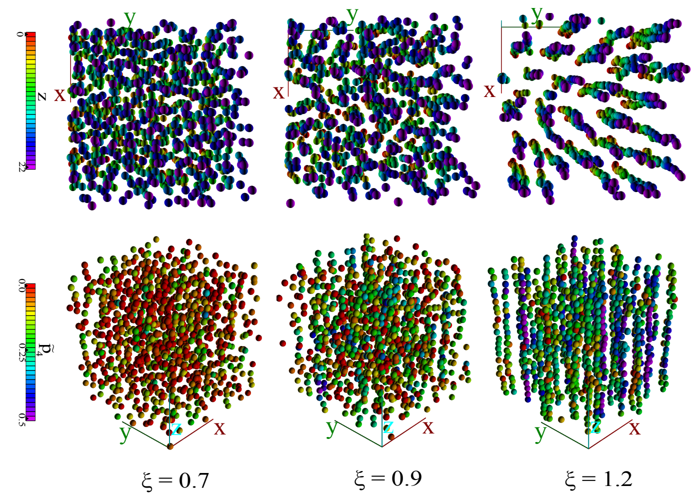
- "a theorist's experiment"... (simple)
- potentials: $\phi_A(x) = \frac{1}{x}^3$ or $\phi_A(x) = \exp(-x^2/\tau)$ or ...
- better visualization:
  (Steinhardt parameters)



particle i, nearest neighbors: $N_{nn}(i)$

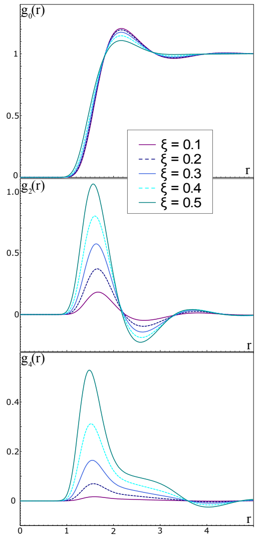
$\tilde{p}_n(i) = \frac{2\pi}{(2n+1)N_{nn}(i)} \sum_{j=1}^{N_{nn}(i)} P_n(\cos\theta_{ij})$

Monte Carlo simulation



$\kappa = 1, \bar{\rho} = 0.1$, 1000 particles, pbc.

Weakly anisotropic fluids



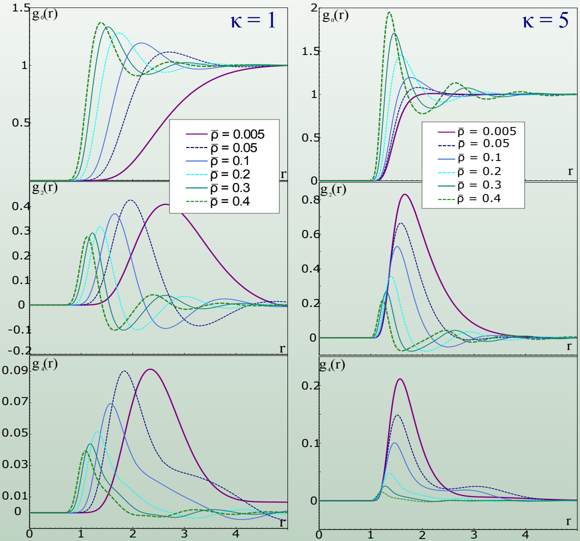
- $\bar{\rho} = 0.1, \kappa = 1, \tau = 6$
- anisotropic correlations (even for tiny ξ)
- effective correlation function

$$g_{\text{eff}}(r) = \frac{1}{4\pi} \int d\Omega \sum_l g_l(r) P_l(\cos\theta) \equiv g_0(r).$$

insensitive for **weakly anisotropic** effects
→direct implications for experiments

- linear in g_2
- nonlinear, but small effect in g_4

Weakly anisotropic fluids (ctd.)

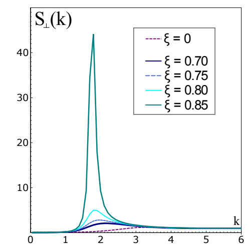


String fluids

OZ approach | Phase transitions

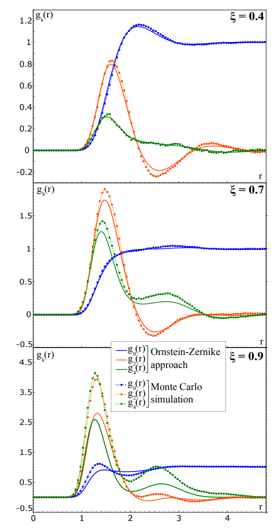
Candidate of PT $\Longleftrightarrow$ Poles of $S(\mathbf{k})$: (poles can be found numerically)

$$[S(\mathbf{k})]^{-1} = 1 - 4\pi\rho\,[\hat{c}_0(k) - \hat{c}_2(k)P_2(\cos\alpha) + \hat{c}_4(k)P_4(\cos\alpha)]\,,$$

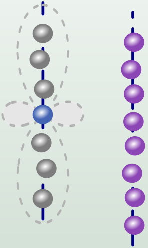


- $\bar{\rho} = 0.1, \kappa = 1, \tau = 6, \xi_{cr} = 0.86$
- divergence of $S(\mathbf{k})$ occurs first for transverse $\mathbf{k}$
- numerical convergence often breaks down before this transition (dep. on τ).

String fluids | MC simulation

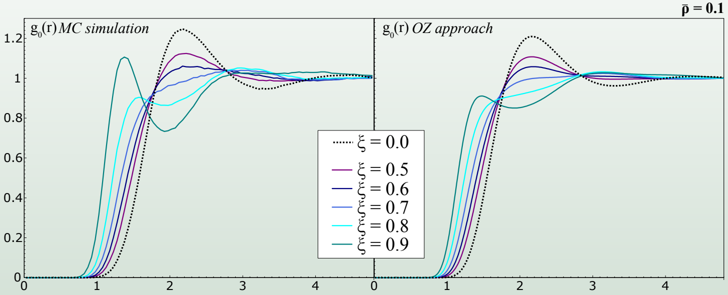


- $\bar{\rho} = 0.1, \kappa = 1, \tau = 6$



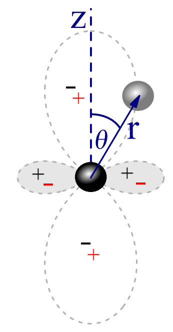
- configurations in string phase:
- interplay g_2, g_4
- typical string-string distance: $r \approx 2.6$
- g_2, g_4 underestimated in string fluid phase

String fluids | Bifurcation of correlation lengths



- g_0 is insensitive in the *weakly* anisotropic phase, but *sensitive* during the transition
- new correlation lengths:
  $\xi_{cr} = 0.86$, (OZ): $\xi_{bifurcation} = 0.84$, (MC): $0.825 < \xi_{bifurcation} < 0.84$
- for some densities: Disappearance of distinct maxima at ξ_{cr}
- experiments?

Sheets | Interaction

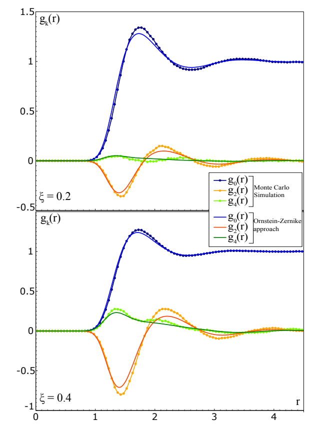


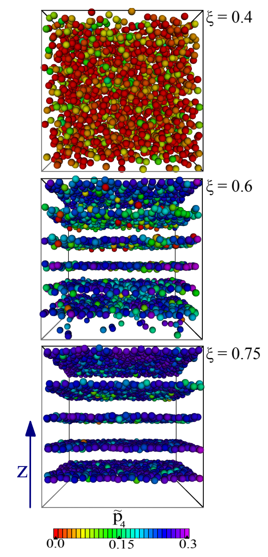
$$V(\mathbf{r}) = \epsilon\left[\phi_{\mathrm{I}}(r) - \xi\phi_{\mathrm{A}}(r)P_2(\cos\theta)\right]$$

- proposed potential in ER-plasmas (R. Kompaneets)
- equiv. to dipolar interaction with $\xi \leq 0$
- attractive and repulsive directions "swapped"
- natural extension of the phase diagram

Sheets

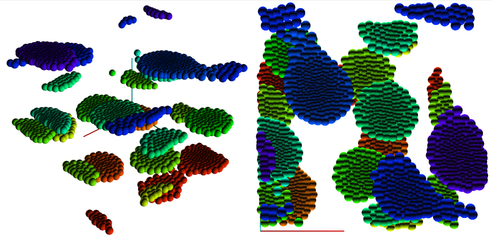
weakly anisotropic fluids positively ERMR:





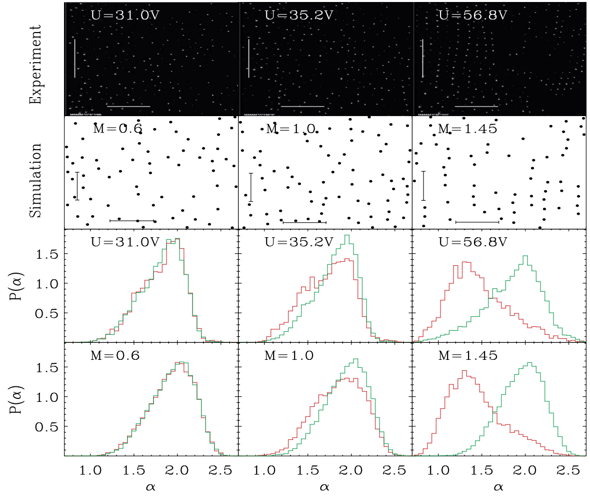
- transition from weakly anisotropic fluids to layers is expected OZ approach at $\xi_{cr} = 0.47$
- $\tilde{p}_4$ calculated for 12 nearest neighbors
- MC simulation: 2000 particles, $\kappa = 1, \tau = 6, \bar{\rho} = 0.1$
- number of sheets per interval (in z-direction) is ξ dependent

Sheets | Mesophases



- first holes and cracks form at $\xi \gtrsim 1.5$
- clearly distinct $\xi \gtrsim 1.75$ "saucer-shaped" particle clusters (mesophase)
- each saucer is itself profoundly planar and perpendicular to z-axes (monolayer/sheets)
- stochastic(?) distribution in z

Experiments | Results (PK-3 plus)



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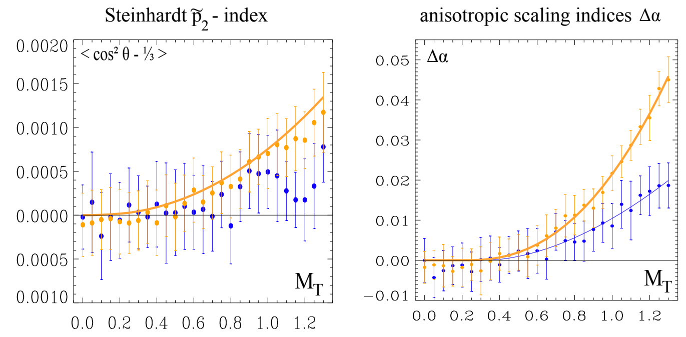
[4] A. Ivlev et al. PRL 100 (2008) 095003

Experiments | Results (PK-3 plus)



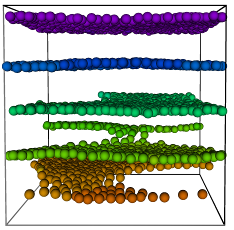
[5]

[5] C. Räth, private communication

Questions

Conclusions

- **weakly** anisotropic fluids
- weakly-to-**string** transition (OZ and MC in good agreement)
- bifurcation in $g_{\mathrm{eff}}(r)$
- pERMR $\longrightarrow$ weakly anisotropic &**sheets**



Open issues

- kinetics of this transitions?
- technological applications? (optical devices?)
- mixtures?
- self-organization?

Thank you for your attention!