UNIVERSITY
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Summary
of Doctoral Thesis

Lāsma Puķina-Slava

INTERFACE SMEARING AND GRAVITY EFFECTS ON MAGNETIC MICRO-CONVECTION

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UNIVERSITY
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FACULTY OF PHYSICS, MATHEMATICS AND OPTOMETRY

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INTERFACE SMEARING AND GRAVITY EFFECTS ON MAGNETIC MICRO-CONVECTION

SUMMARY OF DOCTORAL THESIS

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Subfield: Fluid and gas mechanics

Thesis advisors: Prof. Andrejs Cēbers, Dr. Guntars Kitenbergs

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This thesis is accepted for the commencement of the degree of Doctor of Physics on _____________ 2023 by the Specialized Promotion Council of the scientific section of Physics and Astronomy of the University of Latvia.

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ABSTRACT

In this study an active mixing with a magnetic micro-convection (a finger-like instability) of two miscible fluids water and magnetic fluid is investigated experimentally in microfluidics.

The information about various parameters governing the instability is collected. The influence of magnetic and gravitational fields in the stabilization of the instability is investigated. The results are compared to theoretical predictions done by colleagues.

The results show that the mixing is restricted by the amount of initial smearing and gravity. The shape of the micro-channel and properties of magnetic fluids affect the results. It is demonstrated that the direction of the external magnetic field affects the shape of the instability fingers and the effectiveness of the mixing.

Keywords: Magnetic fluids, Micro-convection, Instabilities, Gravity, Microfluidics

ACKNOWLEDGEMENTS

This dissertation would not have been completed without help and support of my colleagues, family, and friends whom I want to thank here.

I want to thank my thesis advisors Dr. Guntars Kitenbergs and Prof. Andrejs Cēbers who guided this study from its very beginning to the finish. Special thanks to Dr. Guntars Kitenbergs for proposing me this research direction. I also want to thank my colleagues for both practical and emotional support during my PhD studies. I thank Dr. Delphine Talbot (PHENIX lab, Sorbonne University in Paris, France) and Dr. Oksana Petričenko for the magnetic fluids used within this study. Thanks to Dr. Andrejs Tatuļčenkovs for theoretical results, Dr. Ivars Driķis for an Android based synchronization solution for microfluidics pumps, and Michail Maiorov (IPUL) for magnetic measurements. My gratitude goes to Dr. Gökhan Ergin for the opportunity to work with 3D micro-PIV system in Dantec Dynamics, Denmark. Thanks for general support and fruitful discussions to my colleagues: Dr. Viesturs Šints, Dr. Andris P. Stikuts, Dr. Aigars Langins, Dr. Jānis Cīmurs.

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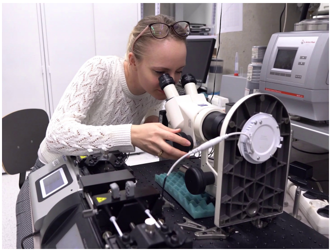
And at last I also want to thank myself for believing in myself and bringing this personal project to completion (see fig. 1.).



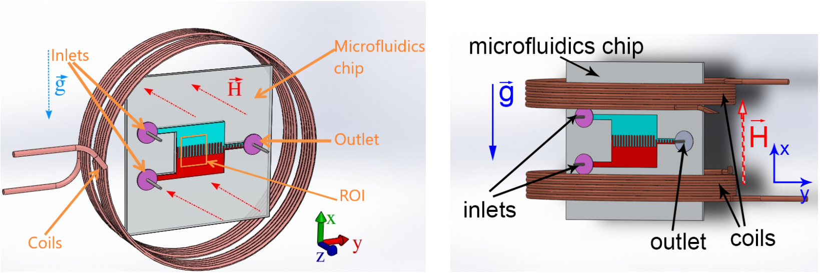
1. Figure: A picture of me looking at magnetic micro-convection

CONTENTS

1. Introduction

1.1. Research actuality and literature review

In this study, two different types of instabilities on the interface between two miscible fluids- water and water based magnetic fluid are investigated experimentally in a vertically placed microfluidics chip in two different magnetic field orientations as showed in figure 1.1.. During the experiments magnetic fluid is placed beneath the water and the experiments are recorded within region of interest (ROI).



(a) Microfluidics chip in a horizontal magnetic field that is perpendicular to the microfluidics chip.

(b) Microfluidics chip in a vertical magnetic field that is parallel to the microfluidics chip.

1.1. Figure: An illustration of the microfluidics chip within an electromagnet

Magnetic fluid is a colloidal solution of magnetic nano-sized particles suspended in a carrier fluid [1, 2]. The size of the particles ensures the colloidal stability against sedimentation. Aggregation of the magnetic particles can be escaped by electrostatic interaction when each magnetic particle have a superficial charge with the same polarity— ionic magnetic fluids [1, 3, 4]. Depending on the purpose of the application of the magnetic fluid the various carrier fluid can be used, for example, water which is convenient biological applications [1]. In this study water based ionic magnetic fluids are investigated, produced with Massart's method [4]. Magnetic fluids produced by this method are stable at a normal pH 7, which together with the fact that they are water based is a requirement for them to be compatible for biomedical applications. Also used maghemite γ-Fe_2O_3 and magnetite Fe_3O_4 particles can provide all of the necessary requirements for application in bio-medicine and bio-engineering [3].

Magnetic fluids possess both magnetic and hydrodynamic properties, therefore making them a compelling research subject [1, 5]. Over the years various applications of magnetic fluids have been created [1, 2, 5–13]. Magnetic fluids respond to an applied magnetic field and modify the external field. At a critical point, there is typically an abrupt change that results in a new equilibrium state. This change often leads to an instability [14]. When exposed to magnetic fields, various instabilities emerge on the interface between magnetic and nonmagnetic fluids [1, 2, 15–27]. Two of these instabilities are broadly investigated in this study on the interface between two miscible fluids. As the characteristic size of the instability is in the scale of microns, these instabilities are termed magnetic micro-convection.

Magnetic micro-convection is a finger-like instability occurring on an interface between magnetic and non-magnetic fluid, caused by a pondermotive force acting on the magnetic fluid in an applied homogeneous magnetic field [14, 28]. The first experimental evidence of the finger-like instability for miscible fluids in a plane layer was delivered already in 1980 by M. Maiorov and A. Cēbers [14]. Though, despite the relatively long time since the phenomenon was first observed the first detailed experimental investigation of the fingering instability caused by the magnetic micro-convection in miscible fluids was carried out only in 2007 by C.-Y. Wen and his colleagues [29, 30], where prominence of the instability fingers were confirmed to be affected by magnetic field strength and the thickness of the microfluidics device. Later it was found out that the magnetic micro-convection also depends on the initial concentration gradient as well as the local viscosity of the mixing interface [31–34]. After the first detailed experimental study with the circular interface between the mixing fluids [29, 30] magnetic micro-convection on a straight interface for miscible fluids was explored in 2008 [35], where the dependence of the instability wavelength and the critical magnetic fields was explored. Extensive study of magnetic microconvection in a horizontally placed Hele-Shaw type microfluidics chip exposed to homogeneous, vertical magnetic field had been carried out by G. Kitenbergs in his Phd thesis [36] in 2015, which was accompanied by several publications [37–39]. There it was discovered that due to the small density difference of the mixing fluids gravity driven convective motion appears, during which a bit denser magnetic fluid slips under the water causing an additional smearing of the interface between the fluids. Although it has been a general assumption that gravity effects can be neglected in microfluidics devices [40] it was disproved during Kitenbergs' research, while showing that gravity can induce a convective flow if miscible fluids have slightly different densities. An attempt was made to measure this convective flow with stereo-μPIV during the research of this dissertation, and the results are presented in the full text of this dissertation. The parasitic gravity driven convective motion can be escaped if thinner channels are used [41] or the microfluidics chip is placed vertically so that a bit denser magnetic fluid is placed beneath the water as it is done in the study of this dissertation, where the effect of gravity by exploring magnetic fluids of various densities and interface smearing between the fluids along with other experimental parameters are explored. Also in this study a different type of instability that emerges if the magnetic field is parallel [17, 42–44] not perpendicular to the microfluidics chip is investigated.

The theoretical model used for the description of the phenomena that is experimentally researched within this study is described by a set of equations, which includes the Brinkman equation, the continuity and convection-diffusion equation [23, 33, 45] and they are reviewed in detail in the full text of this dissertation.

In the study of this dissertation magnetic fluid is considered as an incompressible Newtonian fluid and the viscoelastic contributions are disregarded. Water molecules with the magnetic particles change their their position and direction due to thermal motion, leading to what is referred to as rotational and translational diffusion. [46]. Translational diffusion is reviewed further as it limits magnetic micro-convection [36]. The diffusion coefficient describes the particle's ability move around in a certain time period and it is affected by the size of the particles and the properties of the carrier fluid. The diffusion coefficient can be expressed via Stokes-Einstein equation [47]:

$$D = \frac{k_{\mathrm{B}}T}{3\pi\eta d_{\mathrm{H}}} \tag{1.1}$$

where k_{B} is the Boltzmann constant, T is the temperature, η is the viscosity of the base fluid and d_{H} is the hydrodynamic diameter of a particle [48]. The diffusion coefficient characterizing magnetic particles may be affected by external magnetic field[49], but it is disregarded within this study. For concentration gradient the process of particle diffusion can be described with the second Fick's law [50]:

$$\frac{\partial c}{\partial t} = D\mathit{\Delta} c \tag{1.2}$$

where $\mathit{\Delta}$ is the Laplacian operator, c is the concentration and D is the diffusion coefficient of the diffusing particles and t is for time.

Experimentally within this work magnetic fluid and water are filled in a thin microfluidics chip that can be described by a Hele-Shaw cell. The interface between the fluids are considered to be sharp when they meet and if the fluids are let to mix it is expected that the diffusion of magnetic particles can be observed until the magnetic particle concentration equalizes over whole volume of the mixing fluids. This situation can be described by one-dimensional case of a step-like initial concentration pattern [50]. This is explored more in the full text of this dissertation.

This work is mostly an experimental study, the experimental results are compared to theoretical predictions and numerical simulations carried out by colleagues. The comparison is done both qualitatively by visual materials of the instability as well as quantitatively. To compare the experimental results of this work with the theoretical predictions and numerical simulations carried out by colleagues dimensionless magnetic Rayleigh $\mathrm{Ra_m}$ and gravitational Rayleigh $\mathrm{Ra_g}$ numbers are used. $\mathrm{Ra_m}$ is expressed as a ratio between the characteristic time of the diffusion $\tau_D = h^2/D$ and the characteristic time of motion that is driven by non-homogeneous self-magnetic field of the fluid $\tau_M = 12\eta/M_0^2$, and reads as:

$$\mathrm{Ra}_m = M_0^2 h^2/12\eta D, \tag{1.3}$$

where M_0 is magnetization of the magnetic fluid, h is thickness of the microfluidics chip, η is the viscosity of the fluids and D is the diffusion coefficient. $\mathrm{Ra_g}$ is the ratio between the characteristic time of the diffusion τ_D and the characteristic time of motion due to the gravitational field $\tau_G = 12\eta/\varDelta\rho g h$:

$$\mathrm{Ra}_g = \varDelta\rho g h^3/12\eta D, \tag{1.4}$$

where $\varDelta\rho = \rho_{\mathrm{MF}} - \rho_{\mathrm{H_2O}}$ is the density difference between the denser magnetic fluid below and less dense water above and g is the standard gravity. Thus quantified comparison of the results how the magnetic field is governing and the gravity is stabilizing the magnetic micro-convection is carried out.

As the spatial parameters of the system under the study are in the scale of microns, the term microfluidics is in the title of this work. This leads to the fact that different fields of physics are combined within this study.

Microfluidics is the science of manipulating and controlling fluids of small volumes, where one or all of the spatial dimensions are in the scale of microns [40]. As there are many practical advantages to microfluidical devices, they are widely implemented in various science fields, for example, biotechnology, chemistry, engineering or physics [51–54]. Due to the wide range of multidisciplinary applications of the microfluidics, it has been for some time and still is an active research topic [55–57]. As the geometric scale of the flow is reduced down to microns, the viscous forces increases over the inertial forces. A way to measure the interaction between these two forces is the Reynolds number [40, 58]. Reynolds number Re is a dimensionless parameter defined as a relation between the inertial and viscous forces. For small Reynolds numbers that are typical in microfluidics devices flows usually are laminar and the prevailing mechanism for transport is diffusion. Passive and active mixing can be used as a mechanism for speeding up the mixing of fluids in microfluidics devices [58–60]. If magnetic materials and fields are used, just as in the study of this dissertation, mixing can be conveniently enhanced in a contact-less manner. A part of active mixers that are based on magnetic materials and fields, has opened a sub-field called micro-magnetofluidics or magneto-hydrodynamic active micromixers [59, 61]. One way to accelerate the mixing in microfluidics is by external magnetic field creating instability-magnetic micro-convection [37, 38].

Magnetic micro-convection itself, besides promising applications in the fluid mixing, has aroused an interest as an academic research topic, both experimentally [17, 27] and theoretically [17, 62, 63]. This work was motivated mostly by this academic curiosity to better understand the dynamics of the magnetic micro-convection and the impact of the factors that affect it. A development and an emergence of the magnetic micro-convection instability is set by a mix of various parameters. Over the years several studies have been conducted to quantify the effects of these parameters, however the theoretical branch of the study has been more active. Therefore the detailed study of this dissertation will nicely compliment the experimental research field of the phenomena, as without experimental studies theoretical models can not be verified.

1.2. Objective and research questions

For this study the following goal and tasks have been formulated.

Goal

Investigate how the dynamics and size characteristics of the magnetic micro-convection on the interface between two miscible fluids is affected by various factors- gravity, the strength and intensity of the external magnetic field, the thickness of the already pre-mixed interface between two fluids, the thickness of the micro-channel and the properties of the magnetic fluid.

Hypotheses

During this thesis following hypotheses were formed:

- Micro-convective mixing in horizontal magnetic field will be more effective than mixing in a vertical magnetic field as the fingers of this instability have more active character in a horizontal magnetic field.
- The total mixing length that consists of micro-convective mixing and diffusive mixing can be achieved by a specific value of external magnetic field and does not depend on the thickness of the pre-mixed layer between the fluids.
- The value of the critical magnetic field for a certain amount of the initial smearing of the interface between the mixing fluids is the same value of the magnetic field that must be applied to obtain total mixing

length equal this initial smearing.

Tasks

To achieve the goal of this thesis following tasks were formed:

1. Characterize the existing experimental system for experiments in horizontal, external magnetic field perpendicular to the microfluidics chip. Perform the experiments using this experimental system to observe the magnetic micro-convection in flowing fluids.
2. Improve and modify the experimental system for observing the magnetic micro-convection for initially stagnant fluids in two different orientations of external magnetic field; characterize the experimental system.
3. Perform the experiments to observe the magnetic micro-convection for initially stagnant fluids both in horizontal and vertical external magnetic field.
4. Collect the data of the various factors that govern and characterize magnetic micro-convection during the experiments and write the software code to process this data.
5. Analyze the magnetic micro-convection on a miscible fluid interface. Find out how gravity, pre-mixed interface thickness and the orientation of the external magnetic field affect the mixing efficiency due to the magnetic micro-convection.
6. Compare the experimental results of this work with the theory and numerical simulations done by colleagues as well as give the possible reasons for the differences between these results.

All of these tasks were accomplished. The results are presented in the full text of this dissertation in 4 chapters with complementary information in appendices. The results of this work confirmed the proposed hypotheses.

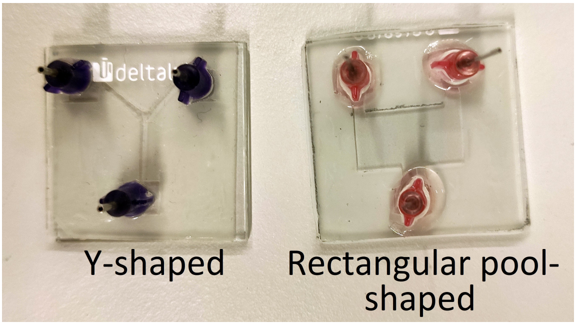
1.3. Experimental methods

The experimental work of this thesis was carried out in the Laboratory of Magnetic Soft Materials (MMML) in University of Latvia in Riga, except for a brief experimental investigation of the fluid flows in a horizontal microfluidics chip with stereo micro-PIV system that was carried out in Dantec Dynamics in Denmark.

In this study magnetic micro-convection with four different magnetic fluids was explored: D107, KTF11-1, KTF09-9 and FF21-5. The magnetic fluid D107 was made in the PHENIX laboratory in Paris, but the other three in the MMML laboratory in Riga. The full list of the magnetic properties and their characterization methods are reviewed in the full text of the dissertation. For some of the magnetic fluids the original magnetic fluid was diluted with distilled water in different ratios to vary the density and particle concentration, in this work this is noted as the "dilution ratio" and the percentage after the name of a particular magnetic fluid describes the fraction of the original magnetic fluid in this magnetic fluid-water dilution.

The microfluidics chips used in this study are mostly made from two microscope cover glasses separated with Parafilm M® spacer. Parafilm M® chips were made with three different thicknesses: $h_1 = 0.135 \pm 0.005$ mm, consisting of one Parafilm M® layer, $h_2 = 0.257 \pm 0.025$ mm and $h_3 = 0.399 \pm 0.015$ mm consisting of two and three Parafilm M® layers accordingly. Microfluidics chips with several different shapes of micro-channels were created. Experiments with continuous fluid flow described in §2.1. were carried out in chips with Y-shaped channel. Micro-channel with a wide rectangular pool was used in the rest of the experiments (§2.2., §2.3. and §2.4.). An example of microfluidics chips used in experiments with Y-shaped and "rectangular pool"-shaped micro-channels is demonstrated in figure 1.2.. The production of the microfluidics chips as well as the justification for the chosen micro-channel shapes are reviewed in the full text of this dissertation.

The microfluidics chip is placed vertically. The magnetic micro-convection is induced by the external magnetic field which is either horizontal and perpendicular to the microfluidics chip or vertical and parallel to the microfluidics chip. The concept of the experimental system already existed from the research described in [36], however some improvements were made.

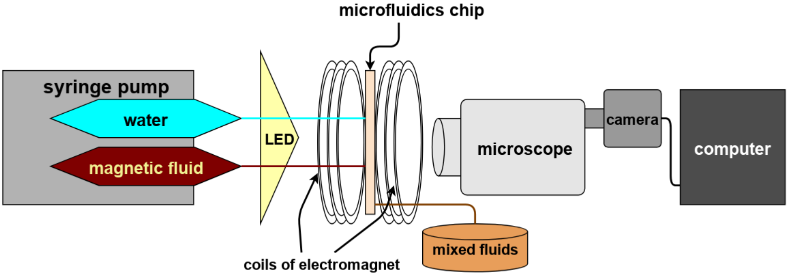


1.2. Figure: Picture of microfluidics chips with the two used micro-channel shapes during this research. On the left Y-shaped micro-channel is visible and on the right micro-channel with rectangular pool is demonstrated.

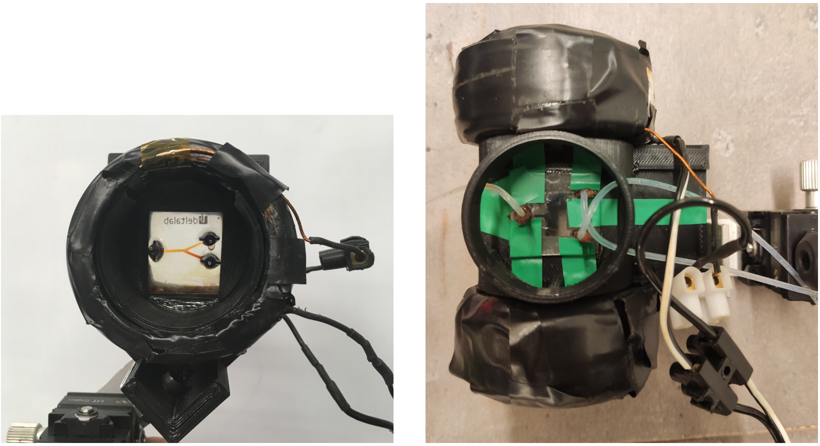
To exclude the gravity caused convective motion the magnetic micro-convection in a vertically placed microfluidics chip is investigated. Conceptually the same experimental system with three small variations was used in this study that are described in greater detail in the full text of this dissertation. Simplified schematic illustration oh the experimental system is demonstrated in figure 1.3.. In general experimental system consists of:

- Microfluidics chip and tubing;
- Electromagnet and its holder;
- LED panel: *Visional®, 4 W, 400 lm, 3000 K*;
- Microscope: *Zeis Stemi 2000-C*;
- Camera: *Lumenera Lu165c, 15 Hz*;
- Computer with necessary software;
- One or two syringe pumps: *Harvard Aparatus PHD Ultra* and *KD Scientific Legato 210P*.

Magnetic fluid is denser than water. Therefore its tubing is connected to the lower inlet of the micro-chip, while water is connected to the upper inlet. The microfluidics chip is vertically fixed in the center of the electromagnet by a 3D printed holder. Depending on the type of the experiments carried out two different coil holders are used as showed in figure 1.4.a.



1.3. Figure: A schematic illustration of the experimental setup.

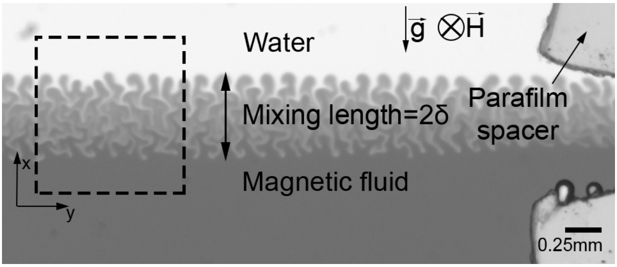


(a) Coils with the holder for experiments with horizontal magnetic field that is perpendicular to the microfluidics chip.

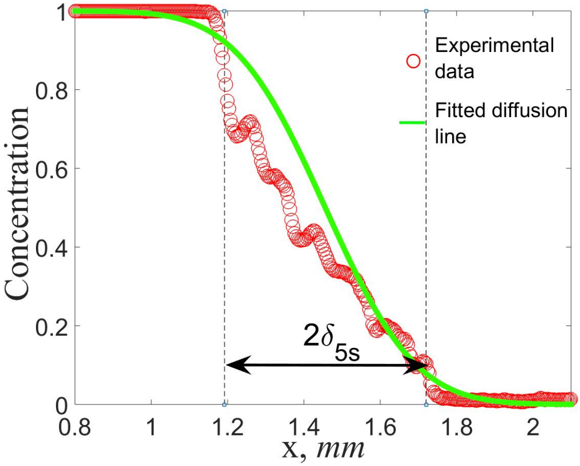
(b) Coils with the holder for experiments with vertical magnetic field that is parallel to the microfluidics chip.

1.4. Figure: Coils of the electromagnet with 3D printed holder and microfluidics chip.

Once the experiment starts the development of the mixing of two fluids is recorded as image series over time with a microscope camera, which is connected to the computer. The data is collected using LabView® based program. The recorded image series are analyzed for a manually selected area- region of interest (ROI) with x and y axes as shown in fig. 1.5.. The mixing length (2δ) characterizes the height of the fingers of the instability. And single δ is diffusion length defined as: $\delta = 2\sqrt{Dt}$.



1.5. Figure: A recorded image of the magnetic micro-convection at $t = 5$ s. Magnetic fluid D107$_{100\%}$; horizontal magnetic field $H = 89$ Oe. ROI is indicated by the dashed line.



1.6. Figure: An average concentration profile (red circles) at $t = 5$ s in an experiment with initially stagnant fluids. Magnetic fluid D107$_{100\%}$; horizontal magnetic field $H = 89$ Oe; micro-channel thickness $h_1 = 0.135$ mm. And the fitted diffusion curve using Fick's law solution (green line). The mixing length 2δ is represented by an arrow.

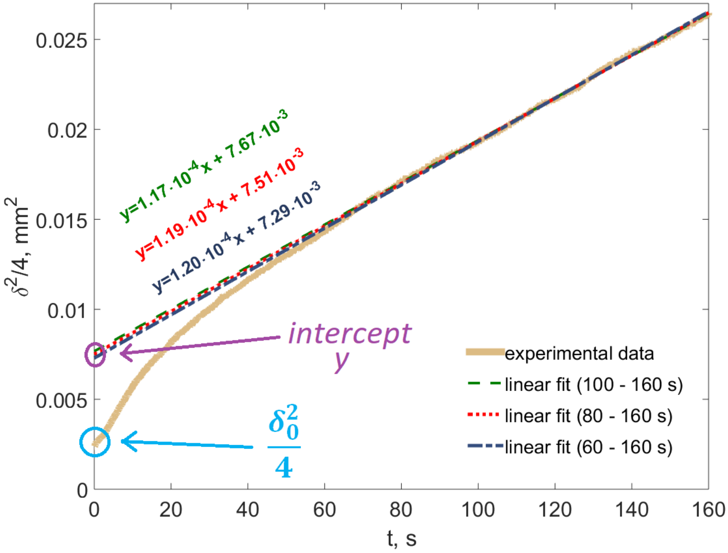
Further the collected data data is processed mostly using MATLAB®

software and the full process is described in detail in the full text if this dissertation. In summary: recorded images as intensity plots are converted to normalized concentration plots via Beer-Lambert law [64, 65]. Then the concentration of each image is averaged along the y-axis and this averaged concentration profile $\bar{c}$(x) is fitted with the diffusion curve, according to Fick's law solution as showed in figure 1.6.. From there the mixing length 2δ is obtained for all of the time-points of each experiment. The equation for diffusion length can be rewritten as $\delta^2/4 = Dt$. Thus the quantity $\frac{\delta^2}{4}$ is directly proportional to the time t the experiment has been happening and the proportionality constant is the particle diffusion coefficient D. To identify the diffusive behaviour it is useful to plot dynamics of the mixing between both fluids as $\frac{\delta^2}{4}$(t). For the experiments in a magnetic field where the micro-convection is present, the line that represents the relationship $\frac{\delta^2}{4}(t)$ has a steep increase at the beginning, but eventually acquires a linear shape. An example can be seen in figure 1.7.. The linearity of the relationship between $\frac{\delta^2}{4}$ and time characterizes the diffusion process, but the initial rise is due to the micro-convective instability. The influence of the micro-convection within this thesis is characterized by a parameter δ_{MC}— mixing length due to magnetic micro-convection. This parameter can be obtained, by considering the function of the mixing dynamics as a sum of both diffusion and micro-convection. In coordinates $\delta^2/4$ with respect to time the initial smearing is visible as the initial rise of the experimental data, as it is demonstrated in figure 1.7. and this value corresponds to $\delta_0^2/4$. Several linear fits are showed in fig. 1.7., they cross vertical axis ($t = 0$) at value y. The mixing due to micro-convection is expressed as $\delta_{\mathrm{MC}} = \sqrt{4y} - \sqrt{4 \cdot \frac{\delta_0^2}{4}}$. So here $\delta_{\mathrm{MC}} \approx 0.07$ mm.

Some of the experiments, mostly the ones carried out in stronger magnetic fields, did not acquire the linear region in coordinates $\frac{\delta^2}{4}(t)$ over the recorded time period. The processing of those experiments is reviewed in the full text of this dissertation. Such experimental results are further referred as results from "attachment method".

Next step of the data processing is information collection about δ_{MC} with respect to various quantities: initial smearing thickness δ_0, external magnetic field H intensity and orientation, various parameters of the magnetic fluids and the thickness of the microfluidics chip h. Also critical magnetic fields H_{c}

are determined for all magnetic fluids used within this research for various δ_0 values. H_c is the value of the external magnetic field at which the instability first emerges.



1.7. Figure: Mixing dynamics between water and magnetic fluid D107$_{100\%}$ in a horizontal magnetic field $H = 56.2$ Oe. Experimental data is shown by a beige line. Other lines are linear fits of the experimental data for different time regions. The thickness of the micro-channel: $h_1 = 0.135$ mm.

Another property that the magnetic micro-convection can be characterized by is the characteristic wavelength λ. It describes the spacial periodicity of the instability. Within the experiments of this study λ was measured once the fingers of the magnetic micro-convection emerged at their base. To compare experiments with theoretical predictions the results are made dimensionless as explained in detail in the full text of this dissertation.

1.4. Approbation of the results

The experimental results of this work have been presented in several international conferences, published in one paper [23], and two other papers that include the results presented in the §2.2. [66] and §2.3. are prepared. Also

Summer School of Complex Fluid-Flows in Microfluidics, which was held at the Faculty of Engineering of the University of Porto in July 2018 was attended during the doctoral studies.

1.4.1. Scientific publications related to the thesis

Part of the results of this dissertation is included in publication about gravity effects on magnetic micro-convection [23]:
G. Kitenbergs, A. Tatuļčenkovs, L. Puķina, A. Cēbers, "Gravity effects on mixing with magnetic micro-convection in microfluidics", *The European Physical Journal E.*, **2018**, *Vol. 41*, pages 138.

These results are included in the main text of this dissertation with kind permission of The European Physical Journal (EPJ).

Another publication that focuses on micro-convective mixing with initially stabilised fluids is prepared [66]:
L. Puķina-Slava, A. Tatuļčenkovs, A. Cēbers, G. Kitenbergs, "How gravity stabilises instability: the case of magnetic micro-convection", *arXiv:2310.15323*, **2023**.

The pre-mixed layer's thickness between the mixing fluids is explored in a publication that is prepared:
L. Puķina-Slava, A. Tatuļčenkovs, A. Cēbers, G. Kitenbergs, "The effects of initially smeared interface on the magnetic micro-convection".

1.4.2. List of scientific seminars and conferences

- L. Puķina, E. Blūms, D. Zablotsky. Nanoparticle transfer in ferrocolloids in nonuniform magnetic field. EuroNanoForum conference. June 21-23, 2017, Valleta, Malta.

- L. Puķina-Slava, A. Tatuļčenkovs, G. Kitenbergs. Experimental investigation of how gravity stabilizes instability of magnetic microconvection. 77th International Conference of the University of Latvia. February 1, 2019, Riga, Latvia.

- L. Puķina-Slava, A. Tatuļčenkovs, G. Kitenbergs. Gravity limited mixing with magnetic micro-convection. International Conference on Magnetic Fluids. July 8-12, 2019, Paris, France.

- G. Kitenbergs, L. Puķina-Slava, A Cēbers. Rivalry of diffusion, external field and gravity in micro-convection of magnetic colloids. International Conference on Magnetic Fluids. July 8-12, 2019, Paris, France.

- L. Puķina-Slava, G. Kitenbergs. Gravity limited mixing with magnetic micro-convection. Seminar of Magnetics and Microhydrodynamics. May 15, 2019, Riga, Latvia.
- L. Puķina-Slava, A. Tatuļčenkovs, G. Kitenbergs. Mixing with magnetic micro-convection bounded by gravity and its effects on pre-mixed fluids. 11th LIQUID MATTER CONFERENCE. July 19-23, 2021, online.
- L. Puķina-Slava, A. Tatuļčenkovs, G. Kitenbergs. Initial interface smearing restricts two fluid mixing with the magnetic micro-convection. 14th European Fluid Mechanics Conference. September 13-16, 2022, Athens, Greece.
- L. Puķina-Slava, A. Tatuļčenkovs, G. Kitenbergs. Pre-mixing effects on magnetic micro-convection. 80th International Conference of the University of Latvia. February 16, 2022, Riga, Latvia.

1.4.3. Research project

A part of this work was supported by ESF project "LU dokturantūras kapacitātes stiprināšana jaunā dokturantūras modeļa ietvarā" No.8.2.2.0/20/I/006.

1.5. Author's contribution

All of the experimental results collected in §2. are obtained and processed by the author of this thesis. The experimental system is manipulated, the microfluidics chips are produced and flow simulation in the microfluidics chip as described in the full text of this dissertation also are done by the author of this thesis. The theoretical predictions and numerical simulations presented in §3. are carried out by colleagues in MMML and found in the literature, but the comparison with the experimental data is carried out by the author of this thesis.

1.6. Author's previous education and scientific experience

Education

- MSc Physics, University of Latvia, 2015.
- BSc Physics, University of Latvia, 2013.

Scientific experience

- Scientific assistant, Laboratory of Magnetic Soft Materials, University of Latvia, *Experimental investigation of magnetic micro-convection.*
- Researcher, Laboratory of Heat and Mass Transfer, Institute of Physics, University of Latvia, *Experimental investigation of magnetic instabilities.*
- Scientific assistant, Optical materials laboratory, Institute of Solid State Physics, University of Latvia, *Spectral investigation of hydroxylapatites.*

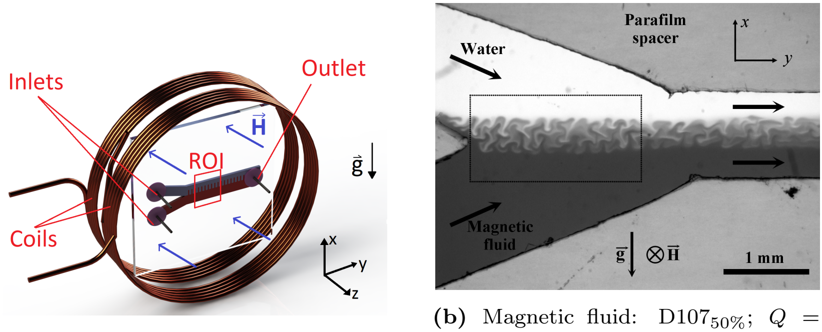
2. Main experimental results

2.1. Magnetic micro-convection in flowing fluids

Within this part of the study microfluidics chips with Y-shaped micro-channel with thickness $h_1 = 0.135 \pm 0.005$ mm were used. One syringe pump was used for pumping in the fluids of interest and the outlet was left open. Microfluidics chip was placed in the center of the coils of the electromagnet so that the magnetic field is perpendicular to the plane of the chip as demonstrated in figure 2.1.. The magnetic field was directed horizontally and perpendicular to the mixrofluidics chip. Part of the results collected in this section are published in publication [23].

In these experiments the density of magnetic fluid D107 was varied using four different dilution ratios (100%, 66%, 50% and 33%) thus exploring gravity effects on the instability. For each dilution the experiments were carried out for different flow-rates set by syringe pump and in different values of the magnetic field.

A desired flow-rate (Q, µl/min) was chosen and the fluids were let to flow for a few moments before the experiment for the interface to stabilize. Then the magnetic field was applied. The experiments are timed and recorded from the moment the magnetic field is switched on. Magnetic field is not changed during all of the experiment. Within short moment after the application of the magnetic field the fingers of the instability emerge across all of the interface. At some time in the experiment the fingers have reached their maximal height and are not growing taller anymore. Fingers have the same height all across the interface. The fingers of the instability emerge earlier if the magnetic field is stronger and also also reach their maximal height faster in stronger magnetic fields. When the maximal height of the fingers have been reached the character of the flow stabilizes. The instability is still continuously forming on the fresh interface on the left side as the flow is going from left to right. And these freshly grown fingers quickly smears on the way to the right side as visible in figure 2.1.b.



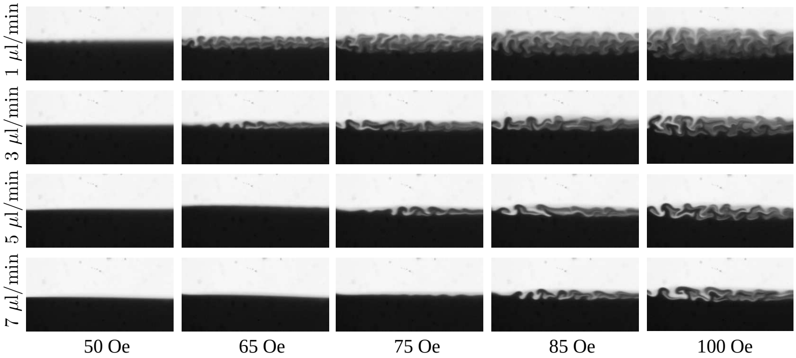
(a) Microfluidics chip in a horizontal magnetic field.

(b) Magnetic fluid: $\mathrm{D107}_{50\%}$; $Q = 1\ \mu\mathrm{l/min}$; $H = 100$ Oe. The dotted rectangle represents ROI. *Fig.2. from [23]*

2.1. Figure: Y-shaped microfluidics chip. Micro-convection is observed while both fluids are being pumped.

Comparing experiments carried out with diluted magnetic fluids, instability emerges later if the magnetic fluid is diluted and the critical magnetic field values for the instability to emerge are higher. Also the maximal height of the fingers is smaller for experiments with more diluted magnetic fluids.

An example of dynamics equilibrium state images of magnetic micro-convection in various external magnetic field strengths for various flow-rates of the fluids is demonstrated in figure 2.2. for magnetic fluid $\mathrm{D107}_{100\%}$ for various magnetic fields. The pattern of magnetic micro-convection emerges only if a sufficient magnetic field, known as the critical magnetic field H_c, is applied. It is visible that the flow-rate affects the critical magnetic field, for higher flow-rates stronger magnetic field must be applied to induce the magnetic micro-convection. One of the desired aims of this study was to find H_c values if both fluids are stagnant at the beginning of the experiment and syringe pump is switched off. During these experiments it was found out that the current setup was not suitable for this, as after the application of the external magnetic field a parasitic flow along the channel (y- direction) appeared and flushed away the fluids. This was resolved later in the next series of experiments with altered experimental setup.



2.2. Figure: Dynamics equilibrium state images of magnetic microconvection for various flow rates and magnetic fields. Magnetic fluid: D107$_{66\%}$. Each image is 1.0 × 2.0 mm large. *Fig.6. from [23]*

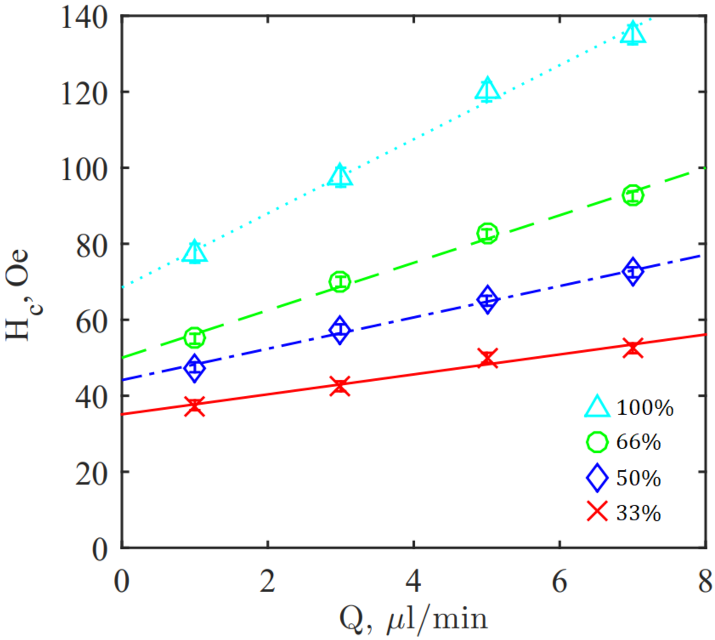
Magnetic fluid	$H_c \pm \Delta H_c$, Oe
D107$_{100\%}$	35 ± 2
D107$_{66\%}$	44 ± 1
D107$_{50\%}$	50 ± 2
D107$_{33\%}$	69 ± 2

2.1. Table: Estimated values of the critical magnetic field with initially stagnant fluids.

Here, the estimation of H_c for initially stagnant fluids was carried out. The critical magnetic field H_c for various flow-rates was measured for all four magnetic fluid dilutions as shown in figure 2.3.. It seems that the critical field can be approximated by a linear relationship depending on the flow-rate, as it is done here. Thus y-intercept at $Q = 0$ μl/min can be found. The critical values for the magnetic fluids used in this part of the study are collected in table 2.1..

Also for all four dilutions of the magnetic fluid D107 the average characteristic wavelength λ for all the measured experiments was found:

- $\overline{\lambda}_{D107_{100\%}} \pm \varDelta\lambda = 0.15 \pm 0.05$ mm;
- $\overline{\lambda}_{D107_{66\%}} \pm \varDelta\lambda = 0.19 \pm 0.03$ mm;
- $\overline{\lambda}_{D107_{50\%}} \pm \varDelta\lambda = 0.22 \pm 0.04$ mm;
- $\overline{\lambda}_{D107_{33\%}} \pm \varDelta\lambda = 0.23 \pm 0.04$ mm.



2.3. Figure: Critical magnetic fields for various flow-rates and different dilutions of magnetic fluid D107. Lines correspond to linear fits that are used to extrapolate the critical magnetic field values at $Q = 0$ μl/min.

The results showed that λ does not depend on the flow-rate of the fluids. It is hard to tell whether the dilution ratio affects the characteristic wavelength of the instability due to the high data dispersion.

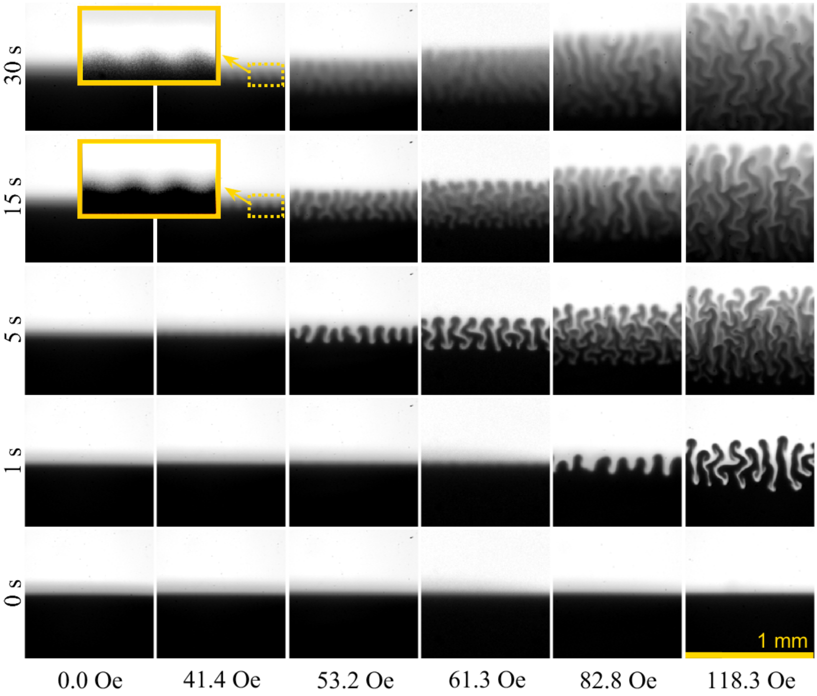
2.2. Magnetic micro-convection in initially stagnant fluids

Experiments described here were carried out with initially stagnant fluids in a vertically placed microfluidics chip with rectangular pool-shaped micro-channel with thickness $h_1 = 0.135$ mm. Here again magnetic fluid D107 with four different dilution ratios (100%, 66%, 50% and 33%) was used. The magnetic field was directed horizontally and perpendicular to the microfluidics chip. The aim of this part of the study was to quantify some parameters of the magnetic micro-convection (critical magnetic field H_c, the characteristic size of the instability λ, mixing due to magnetic micro-convetion δ_MC) and using dimensionless Rayleigh numbers quantify how the gravity restricts this instability. This was done by carrying out experiments without any flow applied from syringe pumps in contrast to the results described in previous section §2.1.. Part of the results reviewed in this section are collected in publication [66].

The timing and recording of an experiment begins when the magnetic field is applied. Before each experiment syringe pumps are used to pump in fresh material in the micro-channel. When a magnetic field higher than H_c is applied, an instability develops across all the interface. The fingers continue to grow until they reach some maximum height. After appearing all fingers of the instability grow at the same rate. Therefore, at a specific time-point all fingers have approximately the same height.

An example of the magnetic micro-convection with initially stagnate fluids is demonstrated in figure 2.4.. First the instability appears as small waves over the interface between the fluids, then small straight fingers appear and if the magnetic field is strong enough they start to bend, curl or even branch. When the fingers have reached their maximal height they still move and swirl but the diffusion slowly takes over as the edges of the fingers start to blur.

The analysis of the finger form with respect to the external magnetic field value and dilution ratio is explored in detail in the full text of this dissertation. The dilution ratio affects the form of the fingers in a sense that stronger magnetic fields must be applied to obtain curled and branched finger forms for more diluted magnetic fluids. The fingers appear earlier and grow out taller in experiments with stronger external magnetic fields and if the magnetic fluid is more concentrated.



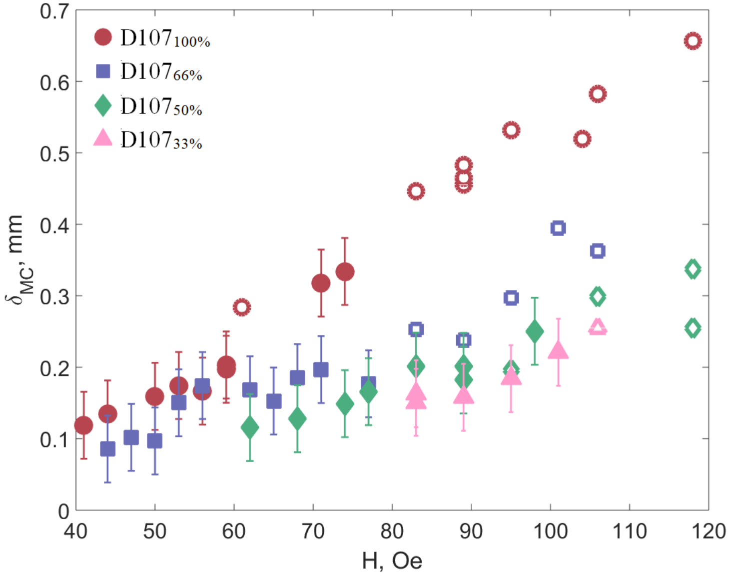
2.4. Figure: Image series of the magnetic micro-convection dynamics with magnetic fluid with D107$_{100\%}$ in various magnetic fields. Each image represents 1×1 mm region.

The experimental dependency of the mixing length due to magnetic micro-convection δ_{MC} on the intensity of the magnetic field H is reviewed in figure 2.5. for all explored dilutions of the magnetic fluid D107.

The average characteristic wavelength of the fingers in the beginning of the instability for experiments within this section was found to be $\overline{\lambda} = 0.12 \pm 0.02$ mm. The critical magnetic field H_c for the instability to emerge was found as well:

- D107$_{100\%}$: $H_c = 41.4 \pm 2.5$ Oe;
- D107$_{66\%}$: $H_c = 44.4 \pm 2.5$ Oe;

- D107$_{50\%}$: $H_c = 59.1 \pm 2.5$ Oe;
- D107$_{33\%}$: $H_c = 79.4 \pm 2.5$ Oe.



2.5. Figure: δ_{MC} with respect to the magnetic field H. The empty markers represent δ_{MC} obtained with "attachment method".

As expected the critical magnetic field is stronger if the magnetic fluid is more diluted. These values are bit higher than estimated from the experiments with flowing fluids (see table 2.1. in §2.1.). It could be due to the fact, that interface between the fluids is sharper if fluids are flowing. As when turning off the syringe pumps a small fluctuation of the interface happens, that sometimes causes small initial smearing. And as it will be reviewed in the next section, interface smearing affects H_c values, as higher H_c are necessary to create an instability if the fluids pre-mixed.

2.3. Magnetic micro-convection in fluids with initially smeared interface between the mixing fluids

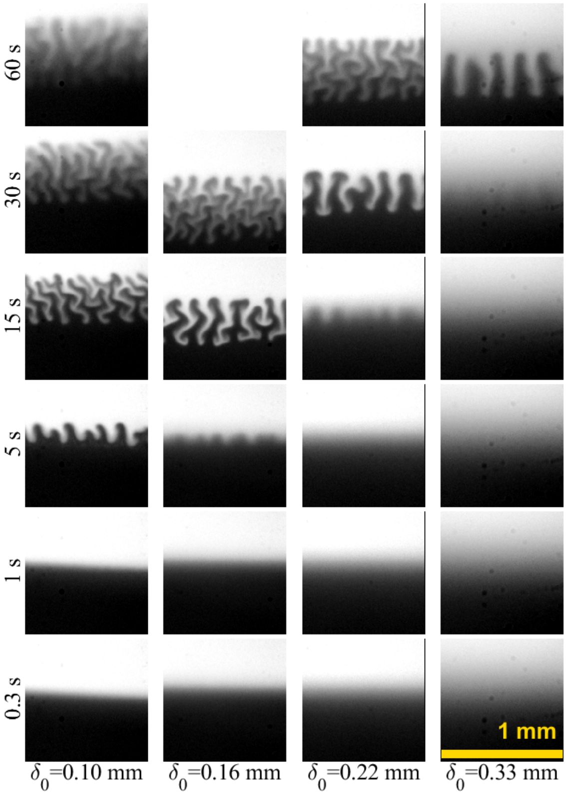
In this section of the work the results of the dynamics of the magnetic micro-convection in experiments with initially smeared interface between the mixing fluids in a horizontal magnetic field are collected. The aim of this part of the study was to find out how the magnetic micro-convection changes if the interface between both fluids is pre-mixed, as it has been showed in the literature [32] that the initial smearing affects the instability. The thickness of the smeared layer δ_0 between both fluids is set and varied by letting the magnetic fluid and water to diffuse over different periods of time before the experiment. The syringe pumps are used to pump in the fresh material for each experiment. An experiment is timed and recorded from the exact moment the magnetic field is applied.

In these experiments three different magnetic fluids were used Similar as in the previous experimental series the magnetic fluids were diluted to various concentrations using distilled water. The diffusion coefficient D is not altered by diluting magnetic fluids, but the particle volume fraction ϕ and density ρ becomes smaller. Therefore the dimensionless quantities $\mathrm{Ra_g}$ and $\mathrm{Ra_m}$ that are used for result comparison are affected as well.

In addition to diluting the magnetic fluids with water the experimental parameters were varied also by using micro-channels with four different thicknesses. By changing the thickness of the micro-channel $\mathrm{Ra_g}$ and $\mathrm{Ra_m}$ also change: $\mathrm{Ra_g} \sim h^3$ and $\mathrm{Ra_m} \sim h^2$.

Visual effects of the initial interface smearing are demonstrated in figure 2.6.. The vertical axis represent the time since the beginning of the experiment (application of the external magnetic field). Horizontal axis represents the information about the initial smearing thickness δ_0 between the mixing fluids at the beginning of the experiment. One column of images represents a single experiment in time. Similar as in previous experiments, the fingers start to grow out straight, then if the magnetic field is high enough they bend and branch. After some time the fingers reach their maximum height and stop growing. After a long enough period of time the edges of fingers become less prominent and they fuse together due to the diffusion.

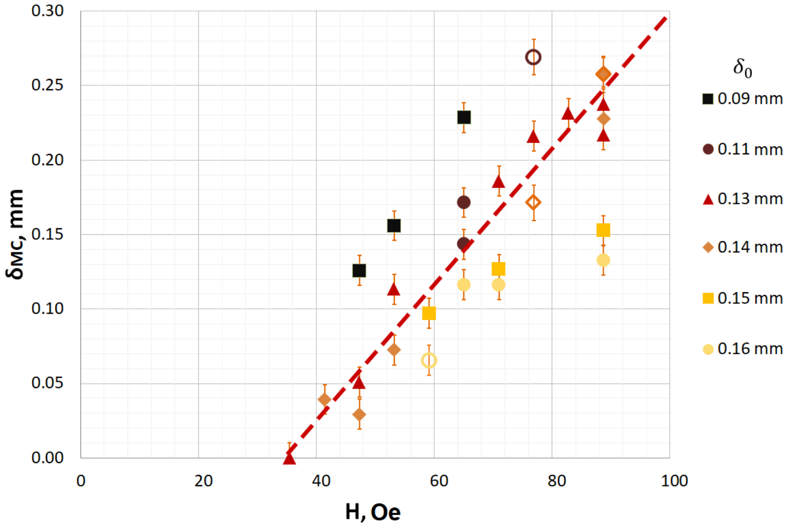
The fingers of the instability appear latter in time and form slower for experiments with higher values of δ_0. The maximum height of the fingers in the same external magnetic field also is smaller in experiments with higher δ_0.



2.6. Figure: Image series of magnetic micro-convection dynamics with different initial smearing thicknesses δ_0 in a constant magnetic field $H = 82.8$ Oe in micro-channel with thickness $h_1 = 0.135$ mm. Magnetic fluid: KTF11-$1_{100\%}$. Size of a region represented by a single image is 1.0×1.0 mm.

It is not conclusive whether the initial smearing thickness affects the characteristic size of the instability. According to [33] the characteristic wavelength λ should increase if the initial smearing thickness δ_0 increases.

For the experiments reviewed in this chapter this is true only for experiments with magnetic fluid KTF11-1, while for other fluids the data dispersion of the results is too large. For the full analysis of the results see the full text of this dissertation. By increasing the thickness of the micro-channel here, the average characteristic λ of the instability increases as well, which agrees with the literature [35].



2.7. Figure: Micro-convective mixing δ_{MC} as a function of a horizontal magnetic field H for various δ_0. Micro-channel thickness: $h_1 = 0.135$ mm, magnetic fluid: FF09-9$_{100\%}$. The empty markers are obtained with attachment method.

The external magnetic field affects the shape of the fingers of the instability, however magnetic field does not affect the characteristic wavelength λ at the beginning of the instability. The value of the magnetic field at which the fingers change their nature was measured and the results are reviewed in the full text of this dissertation. The results showed that the initial smearing affects the form of the fingers. The minimal value of the magnetic field for the fingers to change their character (straight → wavy → branched) is larger both if the magnetic fluid is diluted or if the fluids are pre-mixed. The magnetic field must be stronger for the fingers to change character also if the thickness of the micro-channel is larger.

The quantitative influence of the initial smearing thickness δ_0 on the mixing by magnetic micro-convection δ_{MC} is presented in figure 2.7. for the magnetic fluid FF09-$9_{100\%}$ in a micro-channel with thickness $h_1 = 0.135$ mm. For results with other magnetic fluids and various micro-channel thicknesses please see the full text of this dissertation, where effects of various experimental parameters are compared and explored. Here, the results are grouped by initial smearing thickness and the colors of markers become lighter as the δ_0 increases. Micro-convective mixing is more effective for sharper interfaces (smaller δ_0) between the mixing fluids. The rate at which the mixing δ_{MC} increases by increasing the external magnetic field is not affected by the initial smearing thickness. By extrapolating the data points of a chosen δ_0 until they reach the horizontal axis an approximate critical value of magnetic field H_c for each δ_0 can be obtained. This was done for all experiments, and the results are reviewed in the full text of this dissertation. As expected the likely values of H_{c} appear to be higher for larger δ_0. Additionally, H_{c} for mixing fluids with sharp initial interface ($\delta_0 = 0$) can be estimated from these results. The obtained results of H_c agree within error with the ones collected in previous chapters §2.1. and §2.2..

The results from experiments of this section also show that magnetic micro-convection is oppressed by diluting the magnetic fluid. However, the initial smearing thickness does not affect the rate at which δ_{MC} changes with respect to a different dilution ratio.

Another parameter whose effect on magnetic micro-convection was measured is the thickness of the micro-channel. The largest δ_{MC} values in the same magnetic field with the same initial smearing thickness were achieved in the thickest ($h_3 = 0.399$ mm) micro-channel. However the mixing difference between the two thinner micro-channels ($h_1 = 0.135$ mm and $h_2 = 0.257$ mm) turned out to be statistically insignificant.

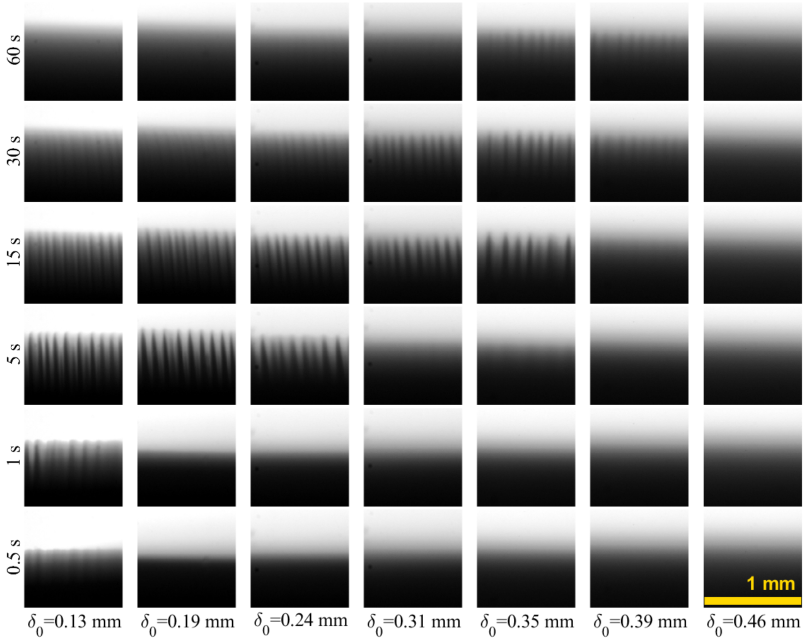
When reviewing the dimensionless experimental results, the dimensionless time parameter t_0 for the same δ_0 also varies between different micro-channels, as the thickness value is used to express $t_0 = \frac{\delta_0^2}{4h^2}$. As expected magnetic micro-convection with respect to $\mathrm{Ra_m}$ is greater for the thinnest micro-channel and therefore the smallest value of $\mathrm{Ra_g}$. The same holds true for more diluted magnetic fluids, as the gravitational effects are decreased.

Within this section of the experiments the validity of the second and third hypothesis proposed in the introduction were explored. The detailed methodology is reviewed in the full text of this dissertation. The results showed that the total mixing length $\varDelta = \delta_{\mathrm{MC}} + \delta_0$ (the sum of the micro-convective and initial smearing lengths) does not depend on the initial smearing thickness, thus confirming the second hypothesis. Also the third hypothesis was confirmed within error: H_c for a certain δ_0 is the same as the magnetic field H to induce δ_{MC} of the same magnitude if $\delta_0 = 0$.

2.4. Magnetic micro-convection in a vertical external magnetic field parallel to the microfluidics chip

In this section of the work the results of the dynamics of the magnetic micro-convection in experiments with the external magnetic field vertical and parallel to the chip are collected. Here, four different magnetic fluids were used. In this section also, the experiments were carried out for several different initial smearing thicknesses in two different micro-channel thicknesses ($h_1 = 0.135$ mm and $h_2 = 0.257$ mm).

Initial smearing effect on the magnetic micro-convection in a constant vertical external magnetic field in the thinnest micro-channel ($h_1 = 0.135$ mm) visually is demonstrated in figure 2.8. in $H = 41.4$ Oe with magnetic fluid FF21-$5_{100\%}$. The vertical axis presents the time since the beginning of the experiment, while the horizontal axis contains information about the initial smearing thickness. One column of images represents a single experiment in time. The periodic pattern of this instability visually differs from the one in a horizontal magnetic field. The fingers are straight and pointy and there is no change in form to wavy and branched fingers unlike in the horizontal magnetic field. Just the same as in the horizontal magnetic field the fingers start to form only if a certain critical value H_c of magnetic field is applied. The fingers form relatively fast and after reaching their maximum height they stop growing. Then over the time the fingers fuse together due to diffusion. The instability appears earlier in experiments with smaller values of δ_0. Also the maximal height of the fingers is reached earlier in experiments with smaller values of δ_0.



2.8. Figure: Magnetic micro-convection dynamics for varioust δ_0 in a constant vertical magnetic field $H = 41.4$ Oe. Micro-channel thickness: $h_1 = 0.135$ mm, magnetic fluid: FF21-5$_{100\%}$. A single image represents 1.0×1.0 mm region.

Within this section values of H_c are determined for various initial smearing thicknesses δ_0. Values of H_c are larger for experiments with larger initial smearing thicknesses δ_0. Also smaller δ_{MC} are achieved for experiments with larger δ_0. Unlike in experiments in a horizontal magnetic field, here the instability appears at the same time, regardless of the strength of the external magnetic field applied.

The effects of micro-channel thickness are also explored here. The micro-convective mixing is more effective in the thickest ($h_2 = 0.257$ mm) micro-channel. Also micro-convective mixing is more effective for more concentrated magnetic fluids, and this difference of δ_{MC} between various dilution ratios is more pronounced in stronger magnetic fields.

Characterization of the wavelength λ of the instability in a vertical magnetic field is not as simple as in the horizontal magnetic field. The fingers appear to form in more than one row along the thickness of the micro-channel. And as the needle-like fingers are searching a stable position the rows of the fingers are sliding pass each other a bit and visually in the pictures it looks like the fingers are becoming thicker and then splitting up. The "thick fingers" are in darker color of black meaning, this formation is less transparent that makes one think, that it might be more than one finger right behind each other. The full analysis of the wavelength for this instability is demonstrated in the full text of this dissertation. Characteristic λ of the instability is not affected by δ_0. The splitting of the fingers has also been observed in literature [17].

Dimensionless analysis shows that same gain of $\mathrm{Ra_m}$ causes greater gain of micro-convective mixing in thinner micro-channel, where gravitational effects $\mathrm{Ra_g}$ are smaller. Just like in experiments with magnetic micro-convection driven by horizontal external field, here as well the gravity suppresses the mixing.

Similar as in a horizontal magnetic field, here as well the value of external magnetic field H is not dependent on the initial smearing thickness δ_0 in order to achieve a specific total mixing length $\mathit{\Delta}$.

2.5. Measurements with stero micro-PIV

Within this study fluid flow in a horizontally placed microfluidics chip with Y-shaped micro-channel was measured using stereo micro Particle Image Velocimetry (PIV) equipment in *Dantec Dynamics* in Denmark. Velocity vectorfield was measured in different horizontal layers of the horizontally placed microfluidics chip at various times since the beginning of the experiment. The detailed review of the results and the experimental setup is visible in the full text of this dissertation. A part of the results agree with intuitively expected motion, where the magnetic fluid slips under the water.

3. Discussion

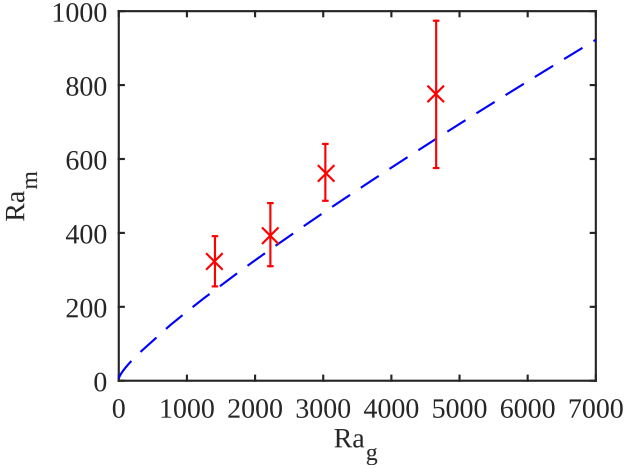
Here, the results of this dissertation are compared with each other. Also numerical simulations and theoretical predictions that were carried out by colleagues are considered.

3.1. Magnetic micro-convection in a horizontal magnetic field

For theoretical considerations the evolution magnetic micro-convection between two miscible, initially stagnant fluids in a Hele-Shaw cell was described by Brinkman equation and the continuity and convection-diffusion equations, which are explored in the full text if this dissertation. The linear stability analysis for a step-like concentration distribution on the boundary between the two fluids was carried out in [23]. The theoretical model considered in the article confirmed that the gravitational force stabilizes the magnetic micro-convection, which agrees to the experimental results of this dissertation.

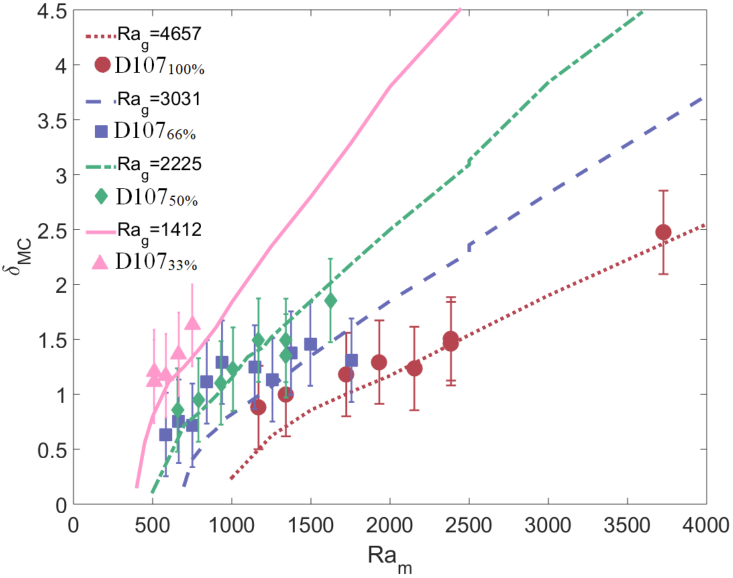
The critical field values H_c estimated from the experiments with flowing fluids for the case with initially stagnant fluids are compared with values obtained from the linear stability analysis in figure 3.1.. As it can be seen the experimental results agree with the theoretical ones within the margin of error. Also this graph confirms the gravitational influence of the instability. For experiments with larger $\mathrm{Ra_g}$ (when the density difference between the mixing fluids is larger) the critical $\mathrm{Ra_m}$ for the instability to first emerge also must be higher. In the full text of this dissertation H_c values obtained numerical simulations as well from experiments with initially stagnant fluids and fluids with initially smeared interfaces are explored in detail. To summarize, all results show good agreement.

Next the results of the experiments with initially stagnant fluids described in §2.2. are compared with theoretical predictions from linear stability analysis and numerical simulations carried out by colleagues. Here the theoretical model (carried out by colleagues) was improved as a system of three liquid layers with isotropic initial magnetic particle concentration was investigated by the linear analysis. This system matches the experimental reality better than the step-like situation with two layers described in [23], as experimentally in the experimental setup used here, there always is a premixed layer of both fluids when starting an experiment. The development of the micro-convection was calculated using Brinkman model, investigating the stability on both borders between two phases of liquids.



3.1. Figure: Comparison of the critical Ra_g and Ra_m between experimental and theoretical results for the instability in a horizontal magnetic field. The blue line represents linear analysis, red markers represent experimental results. *Fig.18. from [23].*

The nature of the micro-convection qualitatively agrees between experiments and numerical simulations. Both in numerical simulations and experiments the fingers of the instability appear after applying magnetic field stronger than the critical one H_c. They grow until they reach some maximal height. After emerging, fingers have uniform height and they tend to bend and branch if the magnetic field is strong enough until the mixing has reached a stage when they remain stationary and only diffusion takes place.



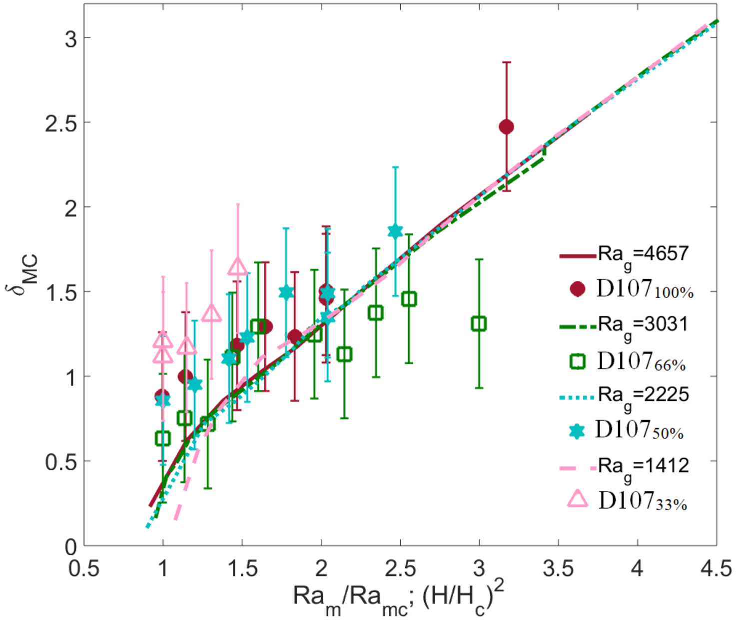
3.2. Figure: Comparison for experimental data (markers) with numerical simulation (lines; carried out by A. Tatuļčenkovs) for δ_{MC} as a function of $\mathrm{Ra_m}$ in a horizontal magnetic field. The case of initially sharp interface.

The comparison of mixing enhancement by the magnetic micro-convection of experimental and numerical data is shown in figure 3.2.. Here the experimental results are converted to dimensionless quantities and expressed using markers. The lines correspond to the results from numerical simulations. The same color is used to describe the experiments with the same gravitational Rayleigh number. For example, the red circles describing experimental results with magnetic fluid $\mathrm{D107}_{100\%}$ correspond to the red dotted line that describes the numerical simulation results for $\mathrm{Ra_g}= 4657$. As the $\mathrm{Ra_m}\sim \chi^2$ experimentally the values of $\mathrm{Ra_m}$ are more restricted for diluted fluids, as by diluting the magnetic fluids its magnetic susceptibility decreases as well. The more diluted the original magnetic fluid, the smaller the value of $\mathrm{Ra_m}$ for the same external magnetic field H. Gravity plays a larger role for the denser (higher $\mathrm{Ra_g}$) fluids, by restricting δ_{MC}.

Also, as explored more in the full text of this dissertation, the comparioson of results show that numerical simulation and experimental results both agree that initial smearing between the mixing fluids restricts the magnetic micro-convection.

The comparison between the experimental and theoretical results of the characteristic finger size of the instability was done as well and is reviewed in detail in the full text of this dissertation. The average wavelength of the instability in experiments with initially stagnant fluids experimentally was found $\overline{\lambda_{\text{stagn}}} = 0.12 \pm 0.02$ mm, for experiments with flowing fluids: $\overline{\lambda_{\text{flow}}} = 0.15 \pm 0.05$ mm, and for initially smeared interface experiments: $\lambda_{\text{smeared}} = 0.16 \pm 0.04$ mm (magnetic fluid D107, micro-channel $h_1 = 0.135$ mm). In experiments with horizontal magnetic chip described in [36] for the same magnetic fluid also $\lambda_{\text{hor}} = 0.15$ mm was found. Recalculating λ obtained from the experiments with initially stagnant fluids to dimensionless wave-number value $\overline{k_{\text{exp}}} = 7.3 \pm 1.0$ is obtained, which agrees with theoretical predictions. All the results from the theoretical model are given considering perfectly sharp interface between the mixing fluids before the application of the magnetic field. However, experimentally it is impossible to create interface that is perfectly sharp. A study in a similar system of magnetic micro-convection [33] has showed analytically that the wave-number k describing the instability decreases if the initial smearing parameter t_0 is increased. The experiments carried out with magnetic fluid KTF11-1 confirmed that λ increases if δ_0 increases. For other magnetic fluids the data dispersion was too big for conclusions.

An interesting development of micro-convective mixing parameter δ_{MC} can be observed if magnetic Rayleigh number Ra_{m} is divided with the critical value $\text{Ra}_{\text{m,c}}$ for numerical simulations and the magnetic field H divided with critical magnetic field H_c for experiments. As it is demonstrated in figure 3.3.. this relationship seems to be coinciding for all values of the used Ra_{g}. $\text{Ra}_{\text{m,c}}$ is proportional to H_c^2. Therefore the ratio H/H_c of the experimental results is squared. The experimental results arrange themselves around the results from the numerical simulations. The experiments are described using markers and lines are used for the numerical simulations. The same color is used to describe results with the same magnetic fluid. The vertical axis of the graph corresponds to the dimensionless mixing parameter due to magnetic micro-convection δ_{MC}. But the horizontal axis describes the ratios $\mathrm{Ra_m}/\mathrm{Ra}_{mc}$ for numerical simulations and $(H/H_c)^2$ for experimental results, and both of these axis coincide. It is suspected that the critical values of the magnetic field and $\mathrm{Ra_m}$ include a significant information about the gravity effects on the development of the micro-convection. Therefore the gravity effect on δ_{MC} is excluded by these divisions.



3.3. Figure: The development of dimensionless δ_{MC}. The numerical results are described with $\mathrm{Ra_m}/\mathrm{Ra_{mc}}$, but the experiments with $(H/H_c)^2$ - both of these axis coincide. The numerical simulations are shown as lines, with the same color as the corresponding experimental markers.

3.2. Magnetic micro-convection in a vertical magnetic field

Experimental results of this dissertation reviewed in §2.4. are compared to studies [17, 37].

The study [37] carried out by K. Ērglis and his colleagues investigates magnetic instability in a horizontally placed microfluidics chip, where external magnetic field is vertical, perpendicular to the microfluidics chip. Experimentally grainy structure was observed there that was caused by a normal field instability as the denser magnetic fluid was slipping underneath water. A theoretical model for the case of a normal-field instability between miscible fluids for a case with infinite vertical direction is investigated there. In [37] characteristic scale of the wave-number is set as $(\Delta\rho/(D\eta))^{1/3}$, where D is a diffusion coefficient, $\Delta\rho$ is a density difference between the mixing fluids, η is the viscosity of the mixing fluids and g is standard gravity. The characteristic wave-number (dimensionless) in [37] is estimated to be $k = (3/32)^{1/3}$. Using these relations the wavelength values λ^* according to the theory carried out in [37] are compared with the experimental ones λ obtained within this thesis in table 3.1.. It must be noted, that the theory in [37] was carried out for infinite layer, while in this thesis the experimental system is confined. Experimental λ values are approximately two times larger, than the ones theoretically estimated. However, experimentally here the primary vivid fingers were counted as explained more in the full text of this dissertation. If the secondary narrow fingers would be included in λ expression, the experimental values of would be even closer to the theoretically estimated λ^*.

	$h_1 = 0.135$ mm		$h_1 = 0.257$ mm				
	D107$_{100\%}$	FF21-5	D107$_{100\%}$	KTF11-1$_{100\%}$	FF09-9$_{100\%}$	FF09-9$_{66\%}$	FF09-9$_{50\%}$
λ, mm	0.09	0.10	0.08	0.11	0.10	0.14	0.12
λ^*, mm	0.05	0.02	0.05	0.04	0.05	0.06	0.06

3.1. Table: Comparison of the wavelength values obtained experimentally and using theory from [37].

The study [17] carried out by M. S. Krakov and his colleagues investigates magnetic instability in a vertical magnetic field, where similar as in the study of this dissertation it was discovered that after the primary peaks have formed, smaller satellites appear between them. The visual splitting of the fingers might appear if the fingers are emerging in several rows as it is explained in in the full text of this dissertation. The finger formation in several rows, although not proposed in [17], seems plausible also due to the size of the characteristic $\lambda \approx 0.1$ mm, as micro-channels with thicknesses $h_1 = 0.135$ mm and $h_2 = 0.257$ mm were used here. Also the study by Krakov *et al.* showed that wavelength of the instability decreases with increasing magnetic field and magnetic Rayleigh number $\mathrm{Ra_m}$. The wavelength determination within this dissertation was inconvenient as the fingers were "shifting", but qualitatively such relationship was not observed here.

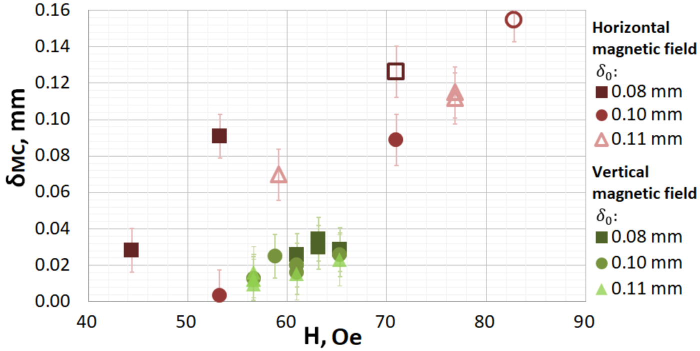
The simulations in [17] also revealed that with an increase in the initial width of the diffusion layer, the wavelength of unstable perturbations increases almost linearly. Therefore it was concluded in the study that the investigated instability is different from the Rosensweig's normal-field instability as then the wavelength of the instability should decrease. However, within the study this dissertation the results show that characteristic λ of the instability is not affected by δ_0.

When comparing the experimental results of this dissertation the first hypothesis proposed in the introduction was confirmed. Greater mixing due to magnetic micro-convection can be achieved in horizontal magnetic field than in vertical one as visible in figures 3.4. and 3.5.. One factor that might enhance the mixing in a horizontal magnetic field, is that fingers in a horizontal magnetic field have more active character.

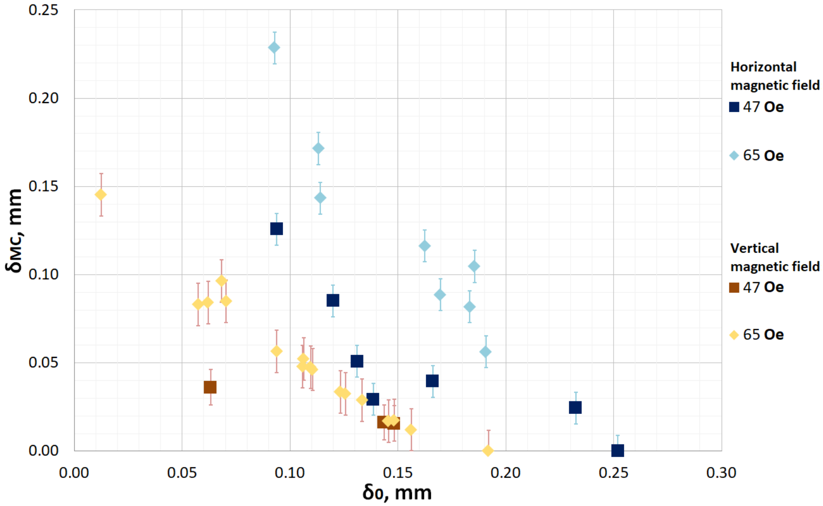
In figure 3.4. micro-convective mixing δ_{MC} with respect to magnetic field H is explored The same δ_0 is represented by the same marker shape. The color of the markers becomes lighter as the value of δ_0 increases. Experiments carried out in a horizontal magnetic field are represented by red markers, while green ones are for experiments carried out in a vertical magnetic field. The rate at which δ_{MC} increases as the value of magnetic field is increased is the same for both directions of magnetic field. Smaller differences of δ_{MC} between two explored directions of the magnetic field were observed in thicker micro-channel ($h_2 = 0.257$ mm).

In figure 3.5. δ_{MC} with respect to the initial smearing thickness δ_0 is explored. The results are grouped by the value of the external magnetic field.

The same value of magnetic field is represented by the same marker shape. The color of the markers becomes lighter as the value of H increases. The experiments in horizontal magnetic field are represented by blue markers, while orange markers are for the experiments in a vertical magnetic field.



3.4. Figure: δ_{MC} in horizontal and vertical magnetic fields H for various δ_0 for KTF11-$1_{100\%}$. Micro-channel: $h_1 = 0.135$ mm.



3.5. Figure: δ_{MC} with respect to δ_0 for various magnetic fields. Micro-channel thickness: $h_1 = 0.135$ mm, magnetic fluid: FF09-$9_{100\%}$.

CONCLUSIONS

Here the conclusions from the experimental results of this dissertation together with corresponding theoretical predictions are arranged in four groups according to the four main experimental sections (§2.1., §2.2., §2.3. and §2.4.) of this work. All three hypothesis proposed at the beginning of this work are qualitatively confirmed. Also, results obtained justify the following **thesis**:

- Magnetic micro-convection can be used to effectively mix fluids in microfluidic devices rapidly, however the area of this mixing is limited by the interaction of various experimental parameters.

Flowing fluids in Y-shaped micro-channel

- Gravitational effects can be important even for microscopic systems with small density differences.
- Gravity and fluid flow stabilize the fingering instability of magnetic-microconvection in a vertical channel: the maximal height of the fingers decreases if the flow-rate of the fluids increases; higher values of critical magnetic Rayleigh number Ra_m are necessary in order to induce the instability for higher values of gravitational Rayleigh numbers Ra_g.
- The experimental results of the critical magnetic field and the characteristic size of the instability were in good agreement with theoretical model of magnetic micro-convection, based on the Brinkman equation.

Initially stagnant fluids

- Parasitic convective motion present when applying horizontal magnetic field present in Y-shaped micro-channels can be escaped, by changing the geometry of micro-channel.
- Gravity limits the micro-convection, by restricting the mixing length due to the micro-convection δ_MC: experiments, numerical simulations, and linear stability results agree.
- Critical magnetic field H_c and magnetic Rayleigh number $\mathrm{Ra}_\mathrm{m,c}$ values portray gravity effects on magnetic micro-convection.

Initially smeared interface between the fluids

- Magnetic micro-convective mixing is restricted by the amount of initial smearing: Experiments and numerical simulations agree.
- Initial smearing restricts the evolution of different forms of micro-convective fingers.
- Initial smearing affects the characteristic size of the instability.
- The characteristic wavelength of the instability is affected by the thickness of the micro-channel: λ increases in thicker micro-channels.
- The total mixing length (micro-convective mixing with diffusive mixing) can be achieved by a specific value of external magnetic field and does not depend on the thickness of the pre-mixed layer between the fluids if the mixing is carried out in a horizontal magnetic field.
- There is an agreement between the magnetic field value to create a certain micro-convective mixing length and the critical magnetic field for a the initial smearing thickness, which is the same as desired convective mixing length with initially sharp interface.
- Critical magnetic field values from various experimental methods are in agreement between themselves and with numerical simulations.

Instability in a vertical magnetic field

- Instability appears at the same time, regardless of the strength of the external magnetic field applied that is higher than the critical magnetic field for the instability to emerge.
- The instability is restricted by gravity: the same gain of $\mathrm{Ra_m}$ causes grater gain of micro-convective mixing in thinner micro-channel, where $\mathrm{Ra_g}$ is smaller.
- Greater mixing due to magnetic micro-convection in the same magnetic field intensity can be achieved in horizontal magnetic field than in vertical one for the same amount of initial smearing between the fluids.
- The characteristic wavelength of the instability explored within this experimental setup is not affected by initial smearing.
- The fingers of the instability split and shift during the experiment in a way that suggests the finger forming in several rows.

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