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On the stability of a free surface of a magnetic fluid under microgravity

S. Odenbach*

Institut für Materialwissenschaften, Bergische Universität GH Wuppertal, Müngstener Str. 10, D-42285 Wuppertal, Germany

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Abstract

We have investigated the behavior of a free surface of a suspension of ferrimagnetic particles in heptane under strongly reduced gravity. It was found that the free surface is destabilized when a homogeneous magnetic field parallel to the surface is applied. The strength of the critical magnetic field parallel to the surface varies with the volume concentration of magnetic particles in the suspension. We show that the destabilization of the fluid layer is forced by the influence of the suspension on the homogeneity of the magnetic field producing a magnetic field component normal to the fluid surface. The dependence of the critical magnetic field on the volume concentration of magnetic particles can be explained by applying the theory of the normal-field instability to this field component under conditions of strongly reduced gravity. The experiments were carried out at the drop tower 'Bremen' of the Center of Applied Space Technology and Microgravity (ZARM) in Bremen.

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1. Introduction

Suspensions of magnetic particles in appropriate carrier liquids are commonly called ferrofluids. The combination of superparamagnetic behavior with normal liquid properties exhibited by these ferrofluids enables the magnetic control of flow and properties of such systems. The suspensions contain magnetic single-domain particles with a mean diameter of about 10 nm covered with a surfactant. The surfactant prevents direct contact between the magnetic particles by steric repulsion and thus the suspension is stabilized against agglomeration of the particles due to van der Waals attraction. By an appropriate choice of the surfactant, numerous magnetic materials like magnetite (Fe_3O_4) or manganese zinc ferrites can be suspended in different carrier liquids like water, heptane or different oils. The magnetic single-domain character of the particles gives rise to the superparamagnetic properties of ferrofluids, which enable a magnetic control of their behavior with magnetic fields in the order of

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* Present address: ZARM, Universität Bremen, Am Fallturm, D-28539 Bremen, Germany.

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50 mT (for further information on ferrofluids, see e.g. Ref. [1, 2]). The combination of this magnetic control with experiments under strongly reduced gravity opens the possibility to investigate hydrodynamic phenomena in ferrofluids without the disturbing effect of gravitational acceleration. This enables e.g. the experimental realization of periodic boundary conditions for convective flows [3]. Such experiments can be performed using microgravity experiment facilities like the drop tower in Bremen.

Focusing on the field of fluid sciences under microgravity, one finds that many experiments require a free surface of the test liquid. Prominent examples are the investigation of Marangoni convection in open containers [4] or free drops [5], the wetting of corners [6] or the behavior of liquid bridges [7]. In these cases, the microgravity environment is used on the one hand to enable the observation of effects suppressed by gravity under normal laboratory conditions or on the other hand to enable the use of large free fluid volumes which cannot be created in ground-based experiments.

It is obvious to consider the use of magnetic fluids for such experiments. For example, to observe the damping of liquid bridge oscillations by magnetic fields, which can be a model for containerless processing of crystals, or for investigation of Marangoni convection driven by magnetic-field-dependent surface tension differences.

In such cases, the question of stability of the free surface cannot only be related to wetting phenomena at the walls of occasionally necessary containers. These effects can be reliably excluded by mechanical or chemical wetting barriers. For a magnetic fluid the influence of a magnetic field in a microgravity environment has to be considered. It is well known that a magnetic field of sufficient strength normal to the free ferrofluid surface will drive a surface instability called normal-field instability [1]. Therefore, only magnetic fields parallel to the free surface will be taken into account in the following.

2. Stability in a magnetic field parallel to the surface

Let us assume a layer of magnetic fluid in a cylindrical container with one free surface as shown in Fig. 1. Wetting of the fluid at the walls of the container is prevented by a sharp edge wetting barrier. The fluid layer is under the influence of a magnetic field parallel to the free surface. This magnetic field has to be sufficiently homogeneous to avoid magnetic forces dragging the fluid over the wetting barriers. For a microgravity environment with a residual acceleration of about $10^{-6}g_0$ ($g_0 = 9.81\ \mathrm{m/s^2}$, normal gravitational acceleration) like it is provided by the drop tower 'Bremen', and wetting barriers with a characteristic length d of about 7 mm, we found magnetic field gradients below $500\ \mathrm{A/m^2}$ to be small enough to fulfill this condition.

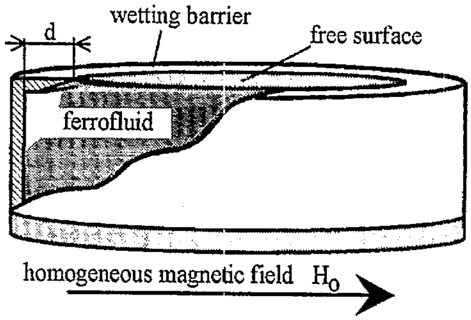

Fig. 1. Schematic view of the fluid layer with free surface and wetting barrier.

The magnetic field induces a magnetization of the magnetic fluid layer. Like it is well known from electrodynamics textbooks (see e.g. Ref. [8]), a magnetized mass in a homogeneous magnetic field will distort the field lines. At the edges of the fluid pool, where the field lines are drawn towards the fluid, the resulting magnetic field will no longer be parallel to the free surface, which means that a small magnetic field component vertical to the free surface of the fluid layer is induced. Fig. 2 shows this component measured at the surface of a cylindrical layer of magnetic fluid 6 cm in diameter and 3 cm thick. As magnetic fluid, a suspension of magnetite particles with a mean diameter of about 10 nm and a volume concentration of 7% in heptane was used.

The layer was under the influence of a magnetic field $H_0 = 5\ \mathrm{kA/m}$ parallel to the top surface. Like it is seen from Fig. 2, the z-component of the field

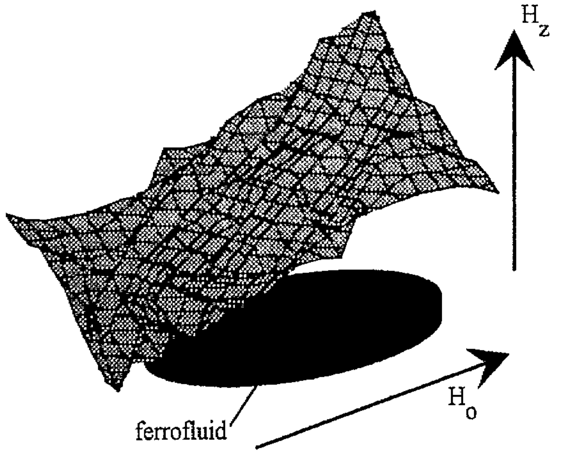

Fig. 2. The component of the magnetic field vertical to the free fluid surface forced by the distortion of the homogeneous magnetic field due to the magnetization of the fluid.

exhibits two extrema. This anisotropy of the field distribution is due to the distinguished direction of the applied magnetic field. Since the reaction of the ferrofluid surface is only determined by the strength of the vertical field component and not by its direction, we can reduce the forthcoming discussion of the stability of the surface to the total value of the maximum of H_z. The dependence of this maximal strength of the vertical component of the magnetic field from the applied field H_0 and from the volume concentration of magnetic particles in the fluid is shown in Fig. 3a and Fig. 3b, respectively. Both relations are linear which can be easily understood by the small-field approximation for the magnetization of the fluid

$$M = \frac{1}{3} M_s \phi \frac{mH}{kT}, \tag{1}$$

where M_s denotes the saturation magnetization of magnetite, ϕ the volume concentration, and m the magnetic moment of the particles, k the Boltzmann's constant and T the absolute temperature of the fluid. Since the vertical field component must vary linearly with the magnetization of the fluid the linear relations in Fig. 3 can directly be seen from Eq. (1).

In principle, any magnetic field component vertical to the fluid surface is able to destabilize the fluid surface if it overcomes a certain critical value given by the theory of normal-field instability. Rosensweig et al. [1] deduced the critical magnetic field strength depending on the fluid properties to be

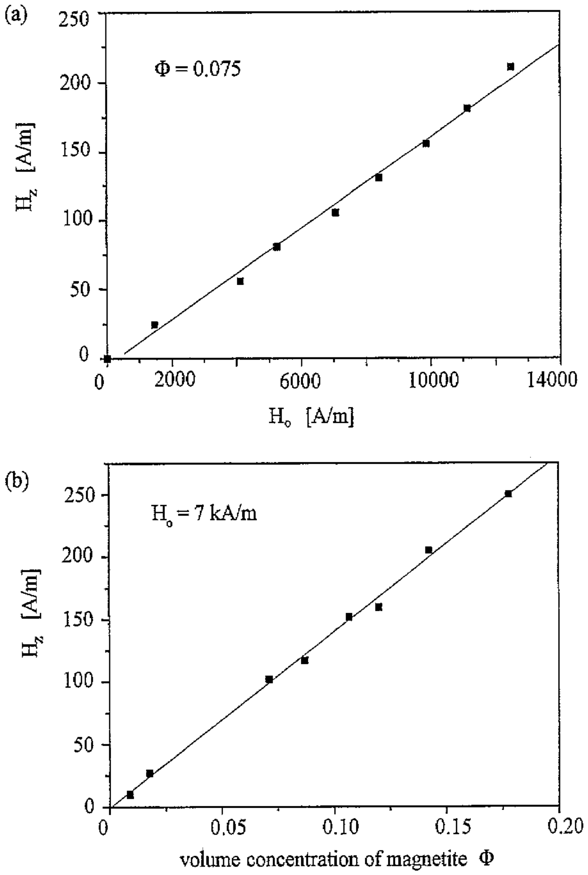

Fig. 3. The dependence of the maximum of the vertical-field component H_z on the strength of H_0 (a) and the volume concentration of magnetic particles (b).

$$H_z^* = \frac{1}{\chi}\left[\frac{2}{\mu_0}\left(\frac{2+\chi}{1+\chi}\right)\right]^{1/2}(\rho g \sigma)^{1/4}. \tag{2}$$

Here σ denotes the surface tension of the fluid, ρ its density and χ its susceptibility (μ_0 is the vacuum permeability). Eq. (2) does not take into account possible influences of a magnetic field parallel to the fluid surface. Such a field should have stabilizing character, as discussed in Ref. [1]. We will show later that this influence can be neglected in our experiments, so Eq. (2) can be used as a basis for the following discussion. For a standard

Table 1
Critical magnetic field strength for various residual accelerations ($g_0 = 9.81$ m/s^2) and fluid H400

g/g_0	H_z^* (kA/m)	Microgravity experiment facility
1	14.1	Ground-based
10^{-2}	4.5	Parabolic flight
10^{-3}	2.5	
10^{-4}	1.4	Sounding rocket
10^{-5}	0.8	Drop tower
5×10^{-6}	0.7	
10^{-6}	0.4	

ferrofluid-like fluid H400 from Table 2, the critical magnetic field strength can be calculated to be of the order of 14 kA/m for normal laboratory conditions. As seen from Eq. (2) the critical field strength scales with $g^{1/4}$ so even weaker magnetic field components vertical to the fluids surface will be able to drive the normal-field instability when gravitational acceleration is reduced. In Table 1 the critical magnetic field strength for the above-mentioned standard ferrofluid is calculated for different residual accelerations in microgravity experiments.

For the experiments with strongly reduced gravity, like they can be performed at the drop tower 'Bremen', the reduction of H_z^* indicates that even the vertical field component produced by the distortion of a homogeneous field parallel to the free surface can be strong enough (see Fig. 3) to destabilize the fluid surface. Therefore, the fluid surface would be able to produce a destabilizing field component even in a highly homogeneous magnetic field. This would give a principle limit for the maximum field strength of a magnetic field parallel to the fluid surface used in microgravity experiments with a free surface of a magnetic fluid. The limit itself would decrease with increasing volume concentration of magnetite in the fluid since the vertical-field component increases with the volume concentration.

3. Microgravity experiments

We have performed microgravity experiments using the drop tower 'Bremen' of ZARM in Bremen to verify the calculations discussed above and to show the existence of an upper limit of the applicable magnetic field strength parallel to the free surface of the magnetic fluid layer.

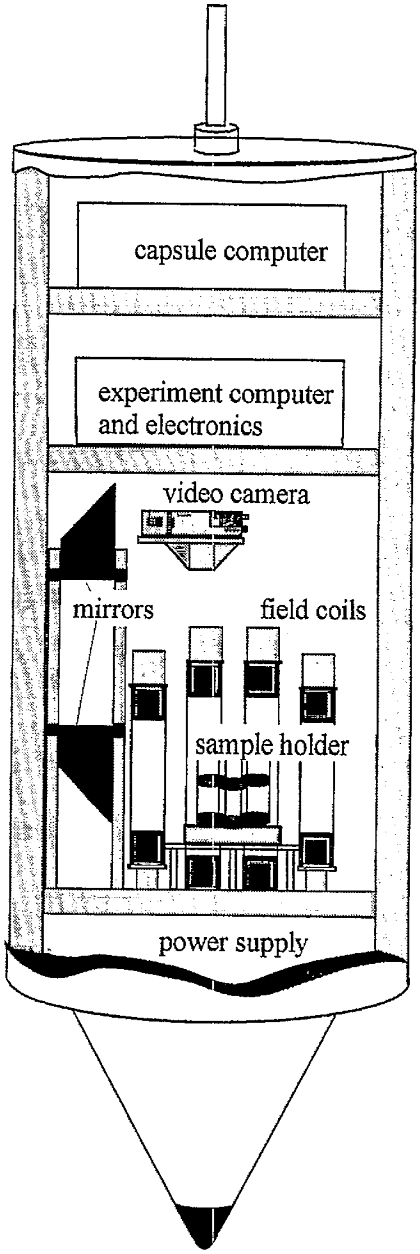

Fig. 4. Schematic view of the experimental setup mounted inside the drop capsule.

The experimental setup shown in Fig. 4 consists of a homogeneous magnetic field produced by

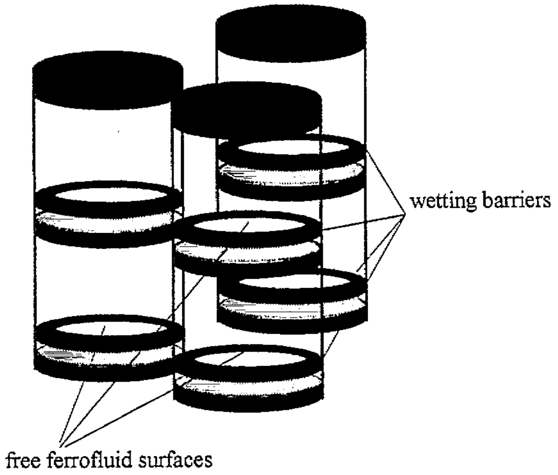

Fig. 5. Schematic view of the fluid containers.

Table 2
Essential properties of the magnetic fluids used in the experiments

Fluid	Symbol in Fig. 6	Φ	χ	ρ (g/cm^3)	σ (10^{-3} Nm)
H50	♥	0.009	0.06	1.00	All Approx. 28
H100	★	0.018	0.13	1.03	
H400	▲	0.071	0.50	1.25	
H500	▼	0.088	0.62	1.31	
H600	◆	0.107	0.75	1.39	
H700	♠	0.124	0.87	1.46	
H800	■	0.143	1.01	1.54	
H1000	●	0.178	1.25	1.68	

four-field coils. The coils are combined in a so-called Fanselau-arrangment giving variations of the magnetic field strength smaller than 0.5% in a sample volume of 15^3 cm^3. The magnetic field is directed vertical to gravitational acceleration and therefore parallel to any free fluid surface brought into the sample volume. The homogeneity of the magnetic field is good enough to suppress effects of magnetic field gradients like they were discussed in Section 1. In the homogeneous region of the coil arrangement, a sample holder is fixed consisting of six cylindrical plexiglass containers. The containers – shown in detail in Fig. 5 – are equipped with a sharp-edge-wetting barrier preventing wetting of the enclosed fluid at the plexiglass walls. The use of six containers enables the investigation of six different magnetic fluids with different volume concentrations of magnetite in one experiment.

The behavior of the fluid surfaces is observed with a video camera giving direct information whether the fluid surface in a certain container is destabilized by the magnetic field under microgravity or not. The whole setup is mounted in the drop capsule of the drop tower 'Bremen'. The capsule provides payload area of about 60 cm in diameter and 1 m high. All functions of the experiment are controlled by an experiment-related computer mounted on a separate platform inside the capsule. This computer provides communication with the capsule computer giving the connection to the control system of the drop tower. For the performance of a microgravity experiment the capsule is dropped in a 120 m high evacuated steel tube providing an experimental time of 4.75 s. For further information on the Bremen drop tower see e.g. Ref. [9].

With the experimental setup described above we have performed a series of 7 drop tower experiments. For each experiment, the containers have been filled with six different magnetic fluids described in Table 2.

From one experiment to the next, the strength of the magnetic field H_0 parallel to the free fluid surface was varied. In that way, the stability of a magnetic fluid surface could be observed as a function of volume concentration of the magnetic component of the fluid as well as of applied magnetic field strength. The magnetic field was switched on 20 ms after the release of the capsule. This time delay is long enough to ensure that the final level of residual acceleration in the order of $10^{-6}g_0$ was reached.

Therefore, it can be excluded that the results are influenced by disturbances of the residual accelerations caused by oscillations of the capsules structure which occurred shortly after its release [10].

Fig. 6 shows the results of the investigations. The different symbols mark the different fluids corresponding to Table 2. Full symbols indicate an unstable fluid surface while open ones stand for

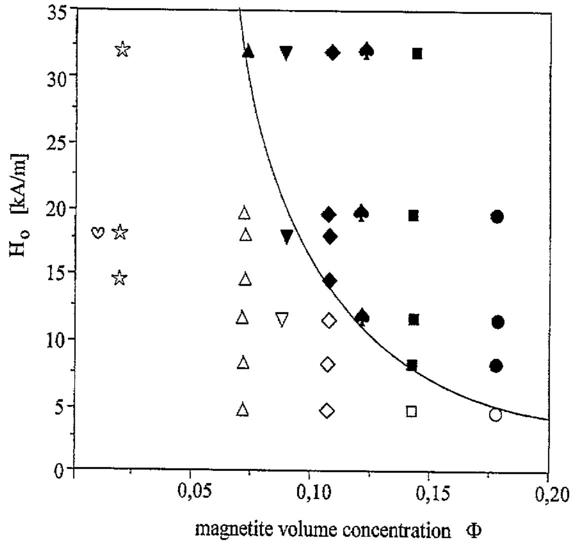

Fig. 6. Measured stability of the free magnetic fluid surface under strongly reduced gravity as a function of magnetite volume concentration and applied magnetic field strength. Full symbols indicate unstable and open ones stable surfaces. The solid line is the border of stability calculated on the basis of the theory for the normal-field instability.

a stable surface. The instability itself is characterized by two fluid spikes located at the positions of the extrema of the magnetic-field component vertical to the fluid surface H_z (see Fig. 2). As is known from the work of Rosensweig [1] the wavelength of the instability increases strongly if gravitational acceleration becomes negligible. In particular, it grows much larger than the dimensions of our fluid pool. Therefore, only one spike at the position of each maximum of the magnetic field strength vertical to the fluid layer can be expected. It was observed that the instabilities occurred with a negligible delay after switching on the magnetic field.

The solid line in Fig. 6, separating stable and unstable regions, can be explained by applying the theory for the normal-field instability to the vertical magnetic field component produced by the distortion of the applied field by the magnetic fluid. In first-order approximation, the critical magnetic field strength $H_z{}^*$ vertical to the fluid surface scales inverse proportional to the susceptibility χ (see Eq. (2)). Since χ is proportional to the volume concentration of magnetite, H_z^* will scale proportional to $1/\Phi$. In addition, we have seen from Fig. 3 that H_z is proportional to the applied magnetic field H_0 and to Φ. Therefore, we find for the critical value of the applied magnetic field

$$H_0^* \propto 1/\Phi^2. \tag{3}$$

In principle, one has to include the influence of variations of volume concentration on the fluids density and the square root term including the susceptibility of the fluid to obtain the margin of stability. The solid line shown in Fig. 6 is calculated including these effects. In the figure, it could not be distinguished from the first-order approximation discussed before. Therefore, we can neglect effects of variations of Φ on the surface tension since they are much smaller than the influences included in the solid line in Fig. 6. In addition, the influence of the magnetic field parallel to the fluid surface, which should have a slight stabilizing effect [1], is also obviously negligible within the measurement accuracy. As seen from the figure, the separation line explains well the behavior of the fluid observed in our microgravity experiments.

4. Conclusion and possible suppression of the destabilization

We have investigated the stability of a free surface of a magnetic fluid subjected to a magnetic field parallel to the fluid surface under strongly reduced gravity. We have found that the fluid itself distorts a homogeneous magnetic field in a way that a destabilizing field component normal to the free surface occurs. It was shown that this field component is able to drive a form of normal-field instability under conditions of strongly reduced gravity. This means that the maximum magnetic field strength applicable to a free magnetic fluid surface is strictly limited by this distortion effect. The dependence of the critical magnetic field strength on the volume concentration of the magnetic component of the fluid could be explained by applying the normal-field theory to the normal field component of the distorted magnetic field. In Fig. 6, it was shown that the theoretical curve taken

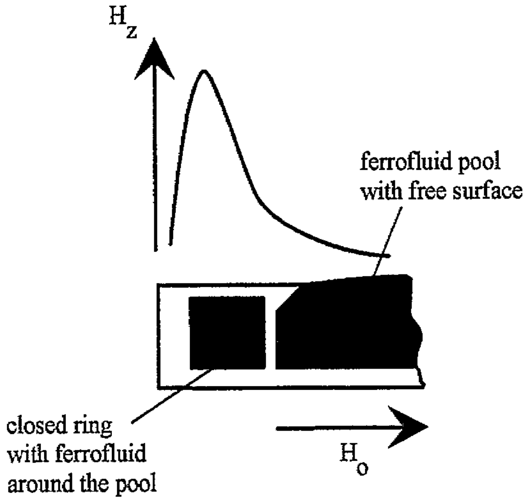

Fig. 7. Principal sketch of the magnetizable boundaries for the free fluid layer.

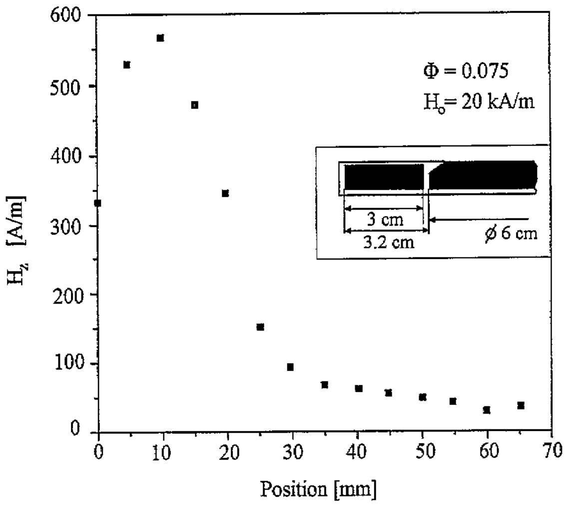

Fig. 8. Measured radial distribution of the normal-field component above the free fluid layer surrounded by a closed ring filled with ferrofluid.

from Eq. (2) represents the experimental data in appropriate form. Therefore, higher effects e.g. the deviation from the linear relation between the normal field component and the applied magnetic field strength, which has to be expected for strong magnetic fields due to the nonlinearity of the Langevin equation had not to be considered.

Beside the fact that we have shown a principal upper limit for magnetic field strength in experiments with free magnetic fluid surfaces alternatives ruling this limit out are conceivable. One possible way out would be the use of microgravity flight facilities with higher residual accelerations. In this case, the critical magnetic field strength would rise as seen from Table 1. For even very strong applied magnetic fields the distortion would not produce vertical magnetic field components strong enough to overcome these values. Nevertheless, if real microgravity with residual accelerations in the order of $10^{-6}g_0$ is requested, or if other flight facilities like sounding rockets are not achievable, the use of magnetizable boundaries for the fluid layer, i.e. a closed ring around the fluid layer filled with the fluid as shown in Fig. 7, could help to stabilize the free surface. In this case, the maximum distortion of the field lines and therefore the maximum distortion-induced normal component of the magnetic field would be shifted away from the free surface.

We have measured the normal-field component in an arrangement with a ring width of 3 cm and a fluid layer with a diameter of 6 cm using fluid H800 (see Table 2) under the influence of an applied magnetic field $H_0 = 20$ kA/m. From the measured radial distribution of the normal-field component shown in Fig. 8 one can see, that its strength in the region of the free fluid layer is too small to destabilize the fluid layer even under drop tower conditions. Since this technique of shifting the maximum of the field disturbances out of the region of the free surface is, in principle, independent of the shape of the pool, it can generally be expected that in this way much higher magnetic fields could be applied without destabilizing the fluid surface.

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