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¹ Authors to whom any correspondence should be addressed.E-mail: egris@science.unideb.hu and bihari.gabor@science.unideb.hu**Keywords:** magnetic spheres, single spheres, chains, self-assemblySupplementary material for this article is available [online](#)**Abstract**

Simple experimental method was developed to examine magnetic self-assembly of macroscopic magnetic spheres of 3mm and 5mm diameters in the lack of external magnetic field. Magnetic force driven aggregation was followed up by video recording and was analysed in detail to identify the processes that lead to the creation of clusters (chains, pairs, circles, etc). Self-aggregation of randomly distributed single spheres, pairs and triplets were examined with this method as well. Applying several liquid media with different viscosity helped to characterize the aggregation processes regarding the kinetic energy of collisions. The results were compared with results of previous experimental works and computer simulations of aggregation of magnetic nanoparticles and magnetic-dipolar systems. Besides to earlier described rings and chains that represent the lowest potential energy of the system of ideal magnetic dipoles in two dimensions, we observed several other structures during the experiments. The most frequently appearing clusters were investigated by direct minimization of the potential energy function of these structures.

Introduction

According to the general definition, self-assembly is the autonomous organization of components into patterns or structures without human intervention [1]. It plays an important role in many practical applications in different fields of physics and technology like creation of novel molecular biomaterials (nanofibres and bionanotubes) [2], and building of nanostructures and nanodevices [3, 4]. It also took place during the preplanetary condensation of the early Solar System, and was observable in magnetic colloids from magnetotactic bacteria [5, 6]. Aggregation of the suspended magnetic particles takes place in the ferrofluids [7–9] and changes the visco-elastic properties of magneto-rheological fluids [10].

Self-assembly of magnetic particles (e.g. cobalt nanocrystals) was examined on nano [11–13] and microscales [14] several times earlier, and the formation of chains and circular structures was reported. Relaxation processes of dipolar magnetic systems towards equilibrium have been investigated either by creating and studying macroscopic scale models [15–17] or using computer simulations [14, 18, 19].

Neodymium magnetic spheres (known as bucky balls or zen magnets) serve as a good macroscopic model of the magnetic particles, because their magnetic field is very similar to the field of the ideal magnetic dipole [20]. Calculations of Messina et al [21] that based on Monte Carlo simulations and direct minimization of the potential energy function had shown, that the minimal energy state realized by different structures according to the number of the constituents. Until three spheres the linear chain, between 4 and 13 spheres the ring form has minimal energy, while above 13 balls, a stacked 3D structure of connected rings creates the minimal energy state. In the case of higher number of dipoles there is an intense and still open discussion about the lowest energy configuration. Instead of the suggested tube-like geometry it was found that for about $N > 1300$ particles a round cluster of densely packed magnetic balls with an fcc lattice is an energetically preferred arrangement [22].

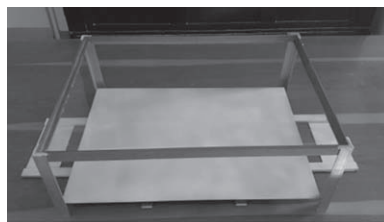


Figure 1. The picture of the experimental arrangement.

According to new findings elongated rod-like structures can even outmatch the structures reported earlier in the cases $N > 460$ [23]. It was suggested recently that in 2D the onion-like structures are preferable for higher numbers of magnetic dipoles [24].

In an earlier experiment of Kun et al. the models of magnetic nanoparticles were small magnet bars attached to larger cork disks, which were floated on the surface of water [15]. The motion of the model particles was described by taking into account the interaction of point like dipoles and by the Stokes drag force exerted by the fluid. It has been found that the structure of the aggregates strongly depends on the initial concentration of the dipoles. The direction of the chain-like structures were oriented by the direction of the Earth's magnetic field.

We were using neodymium magnetic spheres in the experiments, freely moving on a glass plate. In some cases different media were applied to dissipate the kinetic energy. Video recording allowed us to track the formation of chains and closed circular structures, for the detailed understanding of the processes that lead to the creation of these forms. Due to the stronger interaction compared to the earlier experiments, our method was suitable either for studying fast aggregation with large kinetic energy and for observing the effect of fluid resistance. Influence of the Earth's magnetic field was not observable.

Due to the higher kinetic energy we have observed previously unpredicted mechanisms that created wider range of closed clusters than were observed before. The potential energy of these simple clusters was determined by direct minimization of the potential energy of the dipoles as the function of the direction of their magnetic momenta. We suggest, that these clusters represent the local minima of the multi-dimensional potential energy function. In some cases we have found similar magnetic frustration that described earlier [25] as we found different – circular or dipolar – magnetic structures with similarly low potential energies in the case of geometrically identical clusters.

Description of the experiments

We developed a simple device for the experiments. The observed events took place on a thin, $50\text{ cm} \times 70\text{ cm}$ large glass plate with a wooden frame. The glass was glued into the frame, to avoid any leakage of fluid media. Four aluminium legs were holding the framed glass plate at 30 cm high. A closely fitted iron plate was placed under the glass plate. The iron plate was fixed to a wooden grid and two holding arms. In this way, we could reduce the gravitational bend of the plates so, that the gap between the glass and iron plates were less than 1 mm at any point (see figure 1).

We placed neodymium magnetic balls to the glass plate in an even but random distribution. If the balls were at least 3 cm apart from each other, their interaction with the iron plate were stronger than with each other, so the balls were in fixed positions. We have examined the magnetic moments of the fixed balls and experienced that the moments were always vertical – perpendicular to the iron plate – randomly in upward or downward direction. The experiment was started by the sudden drop of the iron plate by which the attraction between the magnetic balls became dominant. The resulting quick aggregation process was filmed at 50 frame per second, so we could not only observe the final results, but the steps of the clustering as well.

In the first series of experiments we did not apply any fluid media in the framed glass plate, so the magnetic balls moved quite fast, against only a slight friction between the balls and the plate.

About 200 pieces of 5 mm large magnetic balls were placed 3–4 cm apart from each other on the glass plate, before dropping the underlying iron plate. The following pictures are showing the course of a typical experiment (figures 2–6, graphs 1 and 2).

After the analysis of the experiments, we have identified several processes of aggregation, and we concluded the following general observations:

During the self-assembly process, the first stage is the formation of dimers by pair-interactions. As the interactions are anisotropic, the movements of the balls towards each other are quite often non-linear. The anisotropic forces are often causing non-central collisions, and thus a large proportion of dimers are created



Figure 2. 0.08 s after the drop/start. There are still solitary, motionless balls. Several cases of pair-interactions and formation of pairs – dimers – are observable. In some cases, non-linear movement of the balls can be noticed. The central collisions are creating slow-moving dimers, while non-central collisions are resulting in quickly rotating dimers. Formation of three ball chains – linear trimers – are also observable by the interaction of dimers and single balls. Some of the trimers are also rotating.

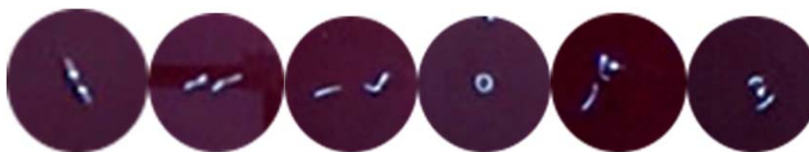


Figure 3. Enlarged parts of the above picture: Central collision, non-central collision, non-linear movement, dimer rotating around the centre of mass, formation of a trimer, rotating trimer. The blurred image of the balls indicates the large speed of them and helps us to assume the magnitude of the speed. The Observing the length of the traces and knowing the time of the exposition the speed can reach up to 0.5 m s^{-1} . The frequency of rotation can reach up to 25–30 Hz.



Figure 4. 0, 18 s after the drop: longer chains of 4–8 balls are forming. Some of the chains are strongly vibrating, wobbling, the blurred picture of some balls suggest large kinetic energy. On the left side in the middle, a 6 ball ring, on the lower right side a 4 ball ring was formed of short chains.

with considerably large angular momentum. These two interesting features – the non-linear movement and the creation of quickly rotating dimers – can be repeated easily with simple manual two ball experiments, without using any equipment.

The next stage of the aggregation process is the interaction of dimers with dimers or single balls. The interaction of dimers with single balls can create rotating or non-rotating trimers during the collisions. The

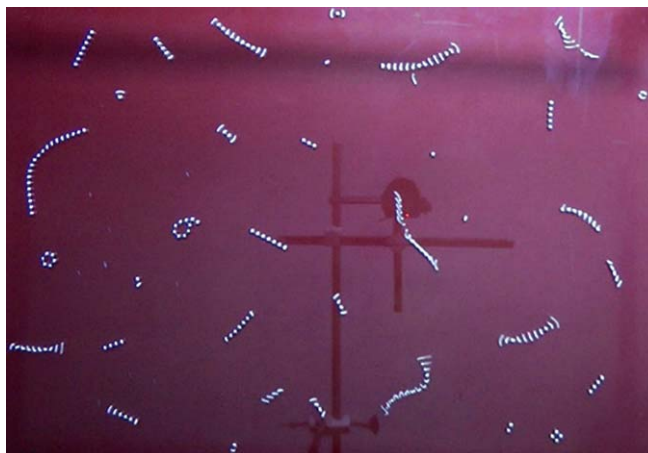


Figure 5. 0, 38 s after the drop: Longer chains are formed by merging of shorter chains and by absorbing the remaining single balls. The collision of single balls into the middle part of a chain causing vibration of large amplitude, which can lead to the joining of the two ends of the chain, thus forming a large ring.

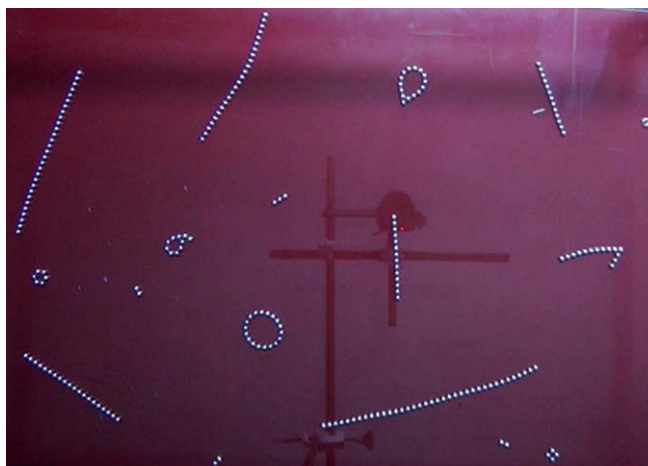


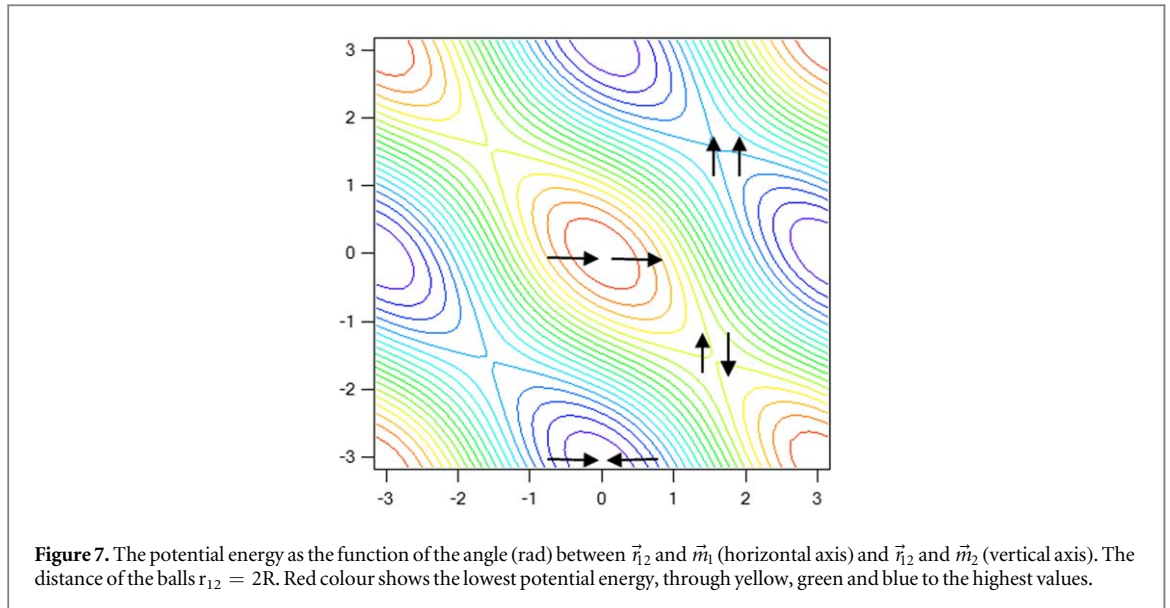
Figure 6. 0, 88 s after the drop: The set run out of single balls, only a few pairs are left – a quickly rotating dimer can be seen in the upper right corner. Beside it, the last single ball is going to impact to a chain. This impact will lead to strong vibration, and the joining of the two ends of the chain, thus forming a D letter shape. The same mechanism formed the two droplet shapes and the broken line shape. The large ring in the middle was formed by the joining of three smaller chains.

dimer-dimer interactions usually creating 4 ball chains. In rare cases two quickly rotating dimers can join each other with opposing magnetic moments, thus forming a 4 ball ring. Theoretical considerations have already proven [14], that this large kinetic energy is necessary to overcome a potential barrier that separates the 4 ball chain from the 4 ball ring.

This process can create 5 or 6 ball rings as well, from the rotating dimers and trimers and single balls. We found that the creation of 5 or 6 ball rings is less probable than the appearance of 4 ball rings. We have seen the formation of a 7 ball ring in this way only once. As the magnetic field of these small rings is basically closed into the ring, these 4–6 ball rings do not interact with other forms on the scene and thus meaning a dead end in the evolution of the aggregation process.

The next stage of the self-assembly is the growth of chains. The shorter chains can join each other or absorb single balls if any remaining. During this stage, single balls are disappearing from the scene. When chains with at least 8 balls are created, two processes can lead to the creation of more complex forms:

1. In some cases, the magnetic moments of linear chains are forming a larger circular structure, and the dragging forces between the ends of the chains can link together these chains, and so, forming large rings. We have observed a case when all the balls on the scene formed one single macroscopic structure.



2. Another unexpected process is observed, that can form rings, broken lines, droplet shapes or D-letter shapes. The initial stage of this process is a single ball, which is going to impact the middle of a chain. A collision at the end of a chain means the simple absorbing of the ball, however, the impact in the middle can cause such a strong vibration, that the two ends of the chain approaching each other, thus starting an attraction and closing the structure.

The impacting ball quite often deforms the line of the balls where it builds into the chain. The breakpoint, created this way is made of three balls, which form an equilateral triangle, forming a 60° angle between the two parts of the chain. This process creates a broken line shape if the kinetic energy is not enough to join the two ends of the chain. With larger kinetic energy, the free ends of the chain join each other, thus forming a droplet shape. In some cases the two sides of the chain have different length, which can cause that the two ends meet again in 60° angle, thus forming another breakpoint and finally a D letter shape.

Due to these processes, the final stage of the aggregation process is the following: Most balls joins long linear chains. The magnetic moments of these long chains often forming large-scale circular structures, but only rarely happens that these linear chains join each other into large rings. Besides the long linear chains, some circles, droplet or D-letter shapes appearing, containing 10–20 balls. Even less frequently, small rings of 4–6 balls are appearing in the picture, usually only one at an experiment including 200 balls.

Theoretical background (figure 7)

The interaction between uniformly magnetized spheres can be analysed theoretically as an interaction between point-like magnetic dipoles, with the limitation that their distance $r_{ij} \geq 2R$, where R is the radius of the solid magnetic spheres [21]. The potential energy of the two sphere system depends on the magnetic moment vectors (\vec{m}_1 , \vec{m}_2) and the relative position vector of the theoretical point-like dipoles at the centre of the spheres (\vec{r}_{12}).

Generally, for the dipoles denoted by i and j [5]:

$$U_{ij} = \frac{\mu_0}{4 \cdot \pi} \left(\frac{\vec{m}_i \cdot \vec{m}_j}{r_{ij}^3} - 3 \cdot \frac{(\vec{m}_i \cdot \vec{r}_{ij}) \cdot (\vec{m}_j \cdot \vec{r}_{ij})}{r_{ij}^5} \right) \quad (1)$$

A simple calculation for two dipoles shows, that in the case of $r_{12} = 2R$ the potential energy has a minimum when \vec{r}_{12} , \vec{m}_1 , \vec{m}_2 are parallel (merging fields). The potential energy function has two inflection points when \vec{m}_1 and \vec{m}_2 are parallel or antiparallel, but perpendicular to \vec{r}_{12} , while the function has an absolute maximum when \vec{m}_1 and \vec{m}_2 are antiparallel and parallel with \vec{r}_{12} as well (opposing fields, see figure 7).

To calculate the potential energy of a multiple ball system, one has to add the potential energies of all possible pair interactions between the balls. Our expectation is that the system evolves to the direction of the energy minimum, so the calculations have to find those forms, where the potential energy per ball value is minimal:

$$U_N = \sum_{i \neq j} U_{ij} \quad \frac{U_N}{N} = \min \quad (2)$$

During the experiments, however there are some limitations, which can cause different final results. Analytical calculations and numeric methods as well can prove that the minimal energy state for two or three dipoles is the

linear chain form, with parallel magnetic moments. (For point-like dipoles in spheres with radius R , on a planar surface) Nevertheless, in the case of 4 or more such dipoles, the closed ring form has the lowest possible potential energy [14]. Between the linear chain form and the ring form, however there is a potential energy barrier, which is the highest for 4 balls, and even though lowering, still considerably high at 8–10 balls. It means that the chains of magnetic dipoles need additional energy to close their ends into circles.

In earlier studies the creation of closed rings were explained by results of direct numeric simulations, where nearby chains with antiparallel polarisation were attached to form closed structures [14]. In our case the interaction between the neodymium magnetic balls was much stronger than in other experiments, therefore we could recognise other evolutionary processes that ended in closed structures. In our experiments for example, a single dipole could impact the side of a chain with such a large momentum, that the transferred kinetic energy was enough for the ends of the chain to overcome the potential barrier and join each other. In the first step of the process, the strike of the impacting ball deforms the chain, and this deformation propagates towards both ends of the chain as a flexible wave. The amplitude of the deformation becomes larger as the wave reaching the free ends of the chain and the kinetic energy of the impacting ball is concentrated onto the last balls of the chain. If the impact reached the chain in the middle, both sides of the chain swing forward at the same time, and at their maximum reach, they can interact with each other, thus closing the chain into a ring.

This mechanism, that closes middle sized chains into rings, have an important effect on the final result of the aggregation process. In the case of such closed ring forms, the magnetic field of the dipoles are concentrated within the structure itself and therefore this structure becomes inactive regarding the magnetic aggregation process. Thus, the appearance of these closed forms can prevent the formation of large-scale structures. Nevertheless, if this closing process becomes less dominant – for example by adding a liquid media to regulate the kinetic energies of balls – than the building up of longer chains can continue. In this case, there is a chance for the chains to form large-scale structures and sometimes all the dipoles on the scene can join each other in one single structure.

Geometric and magnetic structures forming during self-assembly (table 1)

To understand further the self-assembly of these magnetic dipoles, we decided to examine the structures themselves that form as an intermediate or final stage of the process. We used experimental and theoretical methods as well. In the experimental way, we formed manually those sets of dipoles which appeared during the previous aggregation experiments. After the manual creation of chains, rings, droplet or D letter forms, we made the magnetic field of the structure visible by fine iron powder. To avoid the contamination of these strong magnets with the iron powder, first we fixed the structure into plasticine, than covered with a thin glass plate and a paper sheet. Pouring the fine iron powder to the white paper revealed the magnetic field of the structure in detail. Unfortunately, in the immediate vicinity of the structure the magnetic field was usually so strong that the iron powder attracted to the structure, creating a characteristic white, empty zone around it. Even so, the pictures served useful details about these structures.

To examine the structures in a theoretical way, we fixed the structure in a coordinate system. During the calculations we tried to minimize the potential energy function by changing only the direction of the magnetic moments of the dipoles, while the balls had a fixed position as a boundary condition. The calculations are performed by the *IGOR PRO 5.5* program, the optimization was written by the built in function based on the Newton Step Method.

For the better transparency, we have chosen a practical unit for the potential energy (U_0), and the potential energy (U) of every structure is given in this unit. The U_0 unit is the potential energy of two dipoles of which the magnetic moments are parallel to each other but perpendicular to the axis laid through the point-like dipoles.

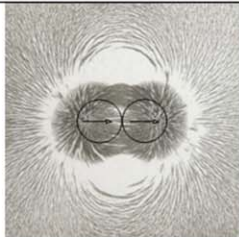
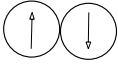

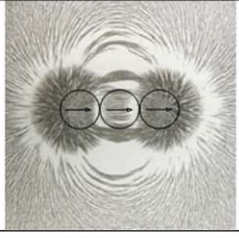
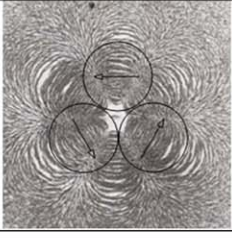
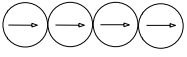
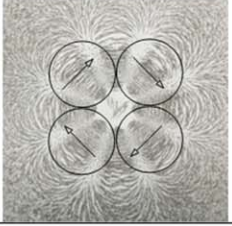
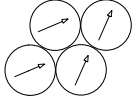
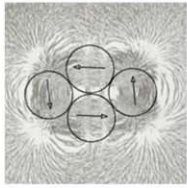
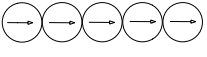
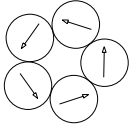
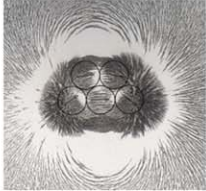
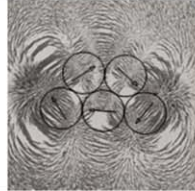
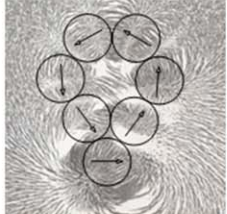
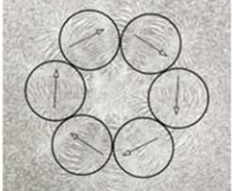
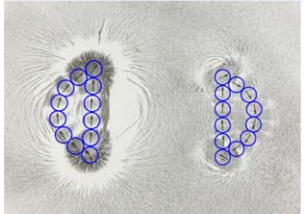
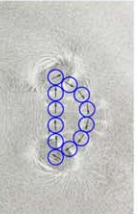
Dimer

In the case of two dipoles, the potential energy is minimal, when \vec{m}_1 , \vec{m}_2 and the axis are all parallel to each other. In this state $U = -1U_0$. This is perfectly in tune with the experiments, as only such dimers with dipolar field could be prepared manually from magnetic balls (See cell D2, table 1).

Magnetic rods or cubes can form another kind of dimer when the magnetic moments are antiparallel. Obviously, the magnetic cubes cannot roll freely as the spheres, so there are potential barriers between the different directions of the moments. Due to this, a stable antiparallel state can exist as a local minimum in the potential energy. Cell C2 shows a couple of two magnetic spheres with antiparallel magnetization. In the case of spheres, the size of the dipoles are always smaller than their distance, so this state is unstable under any circumstances.

This basic distinction between the two low energy states becomes important when examining more complex clusters. The stable dimer form with parallel moments has a dipolar field, while the antiparallel state has a circular field. Our observations suggested, that more complex forms can usually be categorized into these two kinds of structures. Regardless of the geometric form, the structure of the magnetic field better describe the

Table 1. Table of relative potential energies (below) and magnetic structures of the studied clusters. The magnetic field of a single neodymium magnetic ball made visible by iron powder ($R = 25$ mm) can be seen at the upper-right corner of the table.

	D (dipolar field)	C (circular field)	VD (variation of D)	VC (variation of C)
2				
	-1	-0.5		
3				
	-1.42	-1.25		
4				
	-1.64	-1.68	-1.13	-1.5
5				
	-1.79	-1.91	-1.36	-1.73
>5				
	-2.02	-2.05	-2.00	-2.33

clusters, than the geometric form alone. For example, we observed D letter like shapes during self-assembly. But this form can exist with circular and dipolar fields as well. Only the examination of the magnetic field helped to determine, which state created during self-assembly.

Trimer

Three magnetic balls can create two different geometric forms. One of them is linear (See cell D3), while the other is a triangle (See cell C3). The linear form has minimal potential energy if the momenta are parallel to each other and fall to the axis defined by the line of the centers. The dipolar fields of the balls are compensated between the balls, as the north pole of a ball is touching the south pole of the other, so the magnetic field is weaker besides the line of the balls. However the last poles of the chain are not neutralized, so the outburst of the magnetic field can be observed at the two ends of the chain. These two poles are creating a large dipole field, which is similar to that of the magnet bars.

The other possible trimer form is the triangle. This form was scarcely observed during our self-assembly experiments. When shaped manually, this triangle easily ‘jumped out’ to form a linear chain. Theory supports this observation as the linear form has lower potential energy than the triangular form. A similar, triangular shape appears at the breakpoint of larger chains. However, in that case, the magnetic moments set to other directions, so the similarity to the triangular trimer form is only geometric.

Tetramer

Four magnetic balls can arrange in three different shapes in 2 dimensions. The fourth possible shape is the 3-dimensional tetrahedral form, which is very unstable. As for now we are examining two-dimensional self-assembly, we neglect the 3-dimensional forms hereafter.

The linear chain of the four magnetic balls is one of the most common shapes that forms during the first stages of the aggregation process (See cell D4). With its strong dipolar field, it can attract single balls or other chains as well, to create longer chains. The potential energy per ball value decreases as the chains built longer. The theoretical limes of the potential energy for the infinite chain is approximately 2, 404 [21].

Even though the most common form that four balls create during self-assembly was the linear chain, we observed rarely the appearance of the four-ball ring shape as well (See cell C4). This ring form has a slightly lower potential energy than the linear form, however a high potential barrier obstacles its creation. We have never observed that a linear tetramer transforms into a ring – the only possible way is the direct creation of this ring shape from two quickly rotating dimers.

There is a third possible form that four balls can form: a rhomboid shape. It never appeared during self-assembly, and when formed manually, it easily transformed into the rectangular form. This observation was not a surprise though, as this rhomboid form has higher potential energy than that of the rectangular form, so only adhesion and friction can stabilize temporarily this shape.

Our calculations found two possible magnetic structure for the same rhomboid geometry of four balls (Cell VD4, VC4). Manually we could form only the second one, which is still quite unstable.

Pentamer

For five magnetic balls, the minimal energy state is the ring form. Even though there is a considerable energy shift between the linear and circular (See cells D5, C5) forms, we have never seen the transformation between the two – probably due to the above mentioned potential barrier. Five ball rings could only be formed directly by the collision of a rotating dimer and trimer, or two dimers and a single ball.

There is a third possible geometric design for five balls, which has some importance. This is a forerunner of the D letter forms we observed: 3 and 2 balls in two closely fitted chains (Cells VD5, VC5). Although we have never observed such form during self-assembly on its own, this structure can turn up as part of other larger structures, so we examined its properties. The calculations showed that at the minimal energy state the magnetic moments of this structure arrange in a circular form, creating a circular magnetic field. Due to this, when manually creating this state, it spontaneously jumps out to form a ring.

Identical geometry but different magnetic structure characterizes another possible state. In this case, the magnetic momenta are roughly parallel, and thus, this state has a strong, dipole-like field. Even though this magnetic structure has higher potential energy than the circular one, it is stable. So this geometric form can exist only in this higher energy state, as the lower energy state spontaneously reshapes itself into a circle.

Hexamer

During the experiments, we could mostly observe linear, and only rarely circular forms of 6 balls. The linear chains were never transformed into circular forms. The appearing rings were formed by two rotating trimers, and in one case a rotating dimer, a rotating trimer and a single ball joined to create a 6 ball ring. These rare events could happen only in the first step of the aggregation, at a high density of balls.

Compared to the strong dipolar field of the observed linear structures, the small rings have a very weak field outside the circle of the balls (See at the bottom of the table). Thus, it is quite unlikely that a small ring attracts other balls or structures. It means that this form is usually a dead end in the process of aggregation.

Droplet shape

During the experiments droplet-like shapes formed sometimes – less frequently than simple ring forms. Although the shape has usually formed with larger ball numbers, for the sake of simplicity, we indicate here the potential energy of a 7 ball structure. The picture in the bottom-left corner of the table shows the directions of magnetic moments in the minimal energy state. In the case of the manually formed 7 ball ring the potential energy is slightly lower than that of the 7 ball droplet form. As the protruding dipolar area of the droplet shape becomes less dominant when ball numbers are growing, the potential energies of droplets and circles are nearing

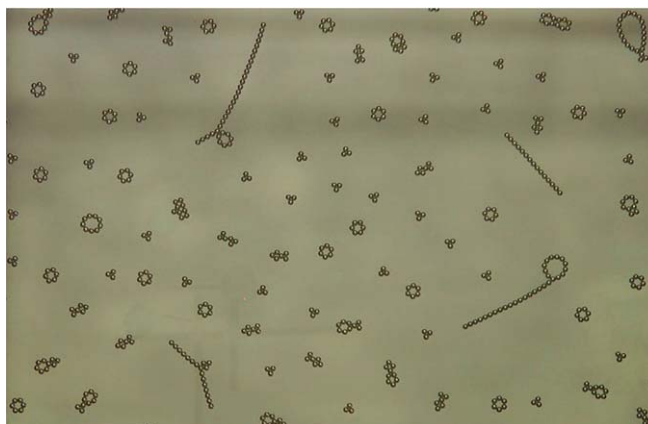


Figure 8. The result of an experiment started with triangular trimers.

each other. The experiments have also shown, that those droplet structures, which has some kinetic energy in the form of vibrations, can easily transform into a ring shape.

D letter shape

The D letter shape is formed when two breakpoints appear on a circular structure. Usually, such form occurs when an impacting single ball hits a larger chain of balls slightly off the middle of the chain. In this way, the first breakpoint forms at the place of the impact, while the other at the meeting point of the two joining ends of the chain. The D letter form is due to the different length of the sides. We observed sometimes the forming of lens shapes with equal side lengths.

Theoretically, there are two different magnetic structure may exist with the same D letter shape. In one of them, the magnetic moments are opposite in the two sides of the D shape. In the other, similarly directed magnetic moments are creating the two sides. In the first case, the magnetic field of the balls is circular, while in the other they form a strong dipole field. Due to the formation process, we expect that only the circularly magnetized form appears in self-assembly. The experiments support this conjecture as we have never observed D letter shape with the dipolar field. See the picture in the table.

According to our observations, in the case of these small clusters static friction between the balls does not plays significant role in the orientation of the magnetic momenta and in the stabilization of the structures. In the case of clean, not painted or eroded metal surfaces static friction forces are much lower than the magnetic forces. In some cases the gradient of the potential energy surface – the force – might be small enough around local minima. In that cases the effect of the friction might be observable.

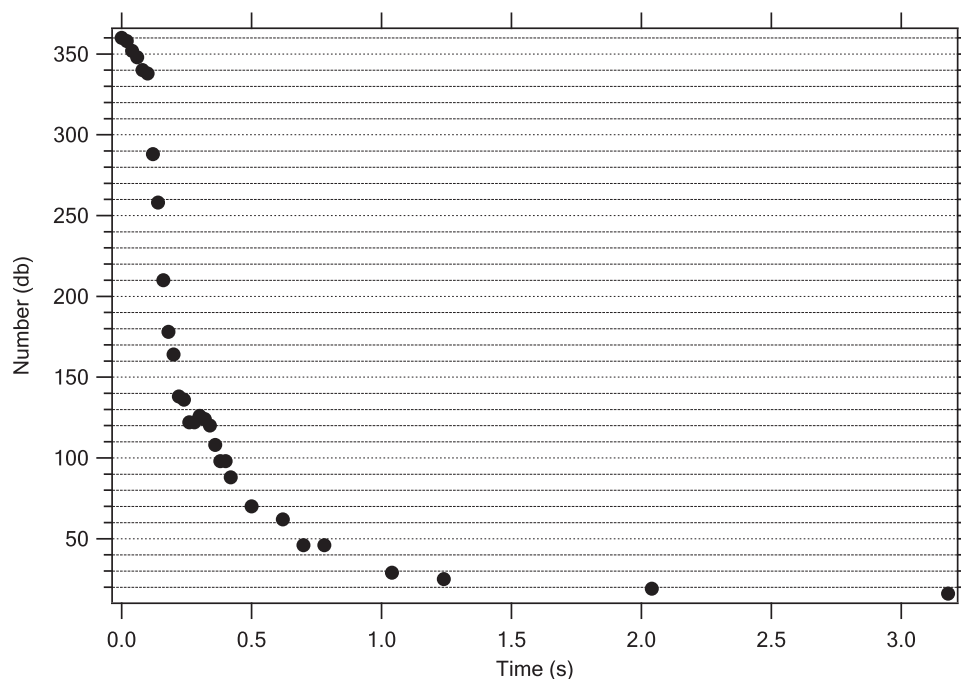
Further experimental developments

Concluding the first series of experiments, we could follow two possible directions to develop the experimental methods further. First, we tried to set a denser distribution of balls on the scene, hoping that it will promote the formation of large-scale structures during aggregation. Nevertheless, it was impossible to increase significantly the distribution density of single balls, as their interaction started to prevail despite the stabilising effect of the iron plate below, and spontaneous chain reactions of self-assembly followed. We tried to solve this problem by distributing dimers and triangular trimers on the plate instead of single balls.

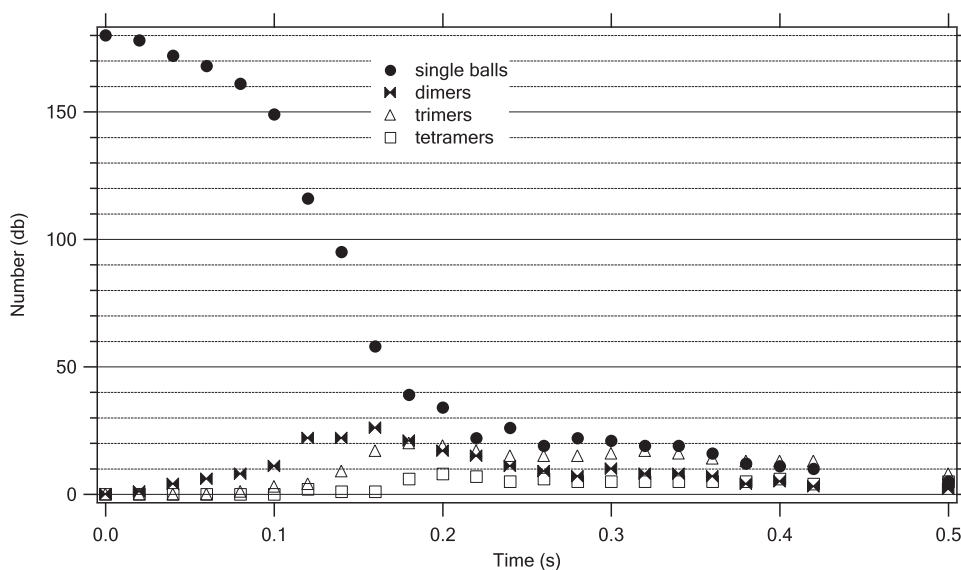
The other direction of development was the application of different liquid media on the glass plate. For this experiment series, the frame was levelled thoroughly and filled so that all balls were covered with the fluid. The most relevant results were observed when we applied water as a low viscosity fluid and very high viscosity silicone oil.

Regarding the first direction of development, the use of dimers increased the density only slightly, as the strong dipolar field of the dimers prevented the creation of a dense distribution. At the same time, the use of dimers simplified the aggregation processes and did not lead us to the observation of new phenomena. The lack of single balls and rapidly rotating dimers decreased the probability for the creation of closed forms. Due to the low levels of kinetic energy, the formation of longer chains was favoured.

Completely different results followed, when we used triangular trimers as the initial state. As these trimers have a relatively weak field outside the triangular structure, they could be set on the plate with quite high density. Figure 8 shows a typical case of the trimer aggregation. Clearly, several trimers did not join the aggregation process, due to their weak attraction. There is a large area in the middle, where the trimers are still intact. Where



Graph 1. shows the number of the sole magnetic poles (chain ends) as the function of the time from the beginning of the aggregation in a typical experiment without applying any media.



Graph 2. shows the number of the chains of $N = 1, 2, 3, 4$ dipoles as the function of the time from the beginning of the aggregation in a typical experiment without applying any media.

the aggregation started, it resulted mostly in various closed forms. The most common shape was the six ball ring, which formed by the simplest interaction of two trimers. However, three or more trimers could also interact simultaneously, which resulted in more complicated forms. The instability of these triangular trimers could also lead to the formation of chains as well: whenever a straightening trimer formed a short chain, the other trimers joined in linear form as well, and built the chain further.

In the other series of experiments, we tried to reduce the kinetic energy of the collisions by the use of liquid media. The use of water slightly reduced the probability of the creation of circular shapes. The medium prevented the long term rotation of dimers and trimers, nevertheless small rings were still appearing. Single balls that impacted into chains were also created circular structures, but with a lesser probability. In this way the aggregation process favoured the creation of long chains and sometimes large structures were formed. Chains of

100 or even more balls created and quite often large scale circular forms appeared. These forms never joined completely into one circular structure, though. Circular forms of 2–3 chains occurred, and once a single U shape is formed from all the balls. But until now, we never experienced the formation of a single ring of all balls.

In another series of experiments, we used silicon oil of extreme large viscosity (AK5000). At the beginning of the considerably slow process, we experienced the formation of short to middle-sized chains by the addition of single balls to the ends of the chains. In the next step, as the single balls disappeared, these middle sized chains turned to each other slowly and joined each other in pairs. However the growth of the chains practically stopped at 10–15 balls length. After a few hours, only one or two new connections formed between the chains, but further built up was hindered by the medium.

We could conclude of these experiments, that the proper use of liquid media could help us to create macrostructures even by simple self-aggregation processes. Thus, we are planning to develop the experiments in this direction.

Summary

During our experiments we have examined the aggregation processes of neodymium magnetic balls with diameters of 3 mm and 5 mm. The sets of balls randomly placed on a glass plate, under which an iron plate fixed the balls. The interaction between the balls became dominant with the sudden drop of the iron plate. The self-assembly of the ball sets was filmed at 50 fps – at this rate the blurred lines helped to estimate the velocity of the balls. The records helped to identify the steps and different ways of the aggregation process. The observations were significantly in tune with the potential energy calculations regardless of the approximations – point-like dipoles and neglected interaction with the surroundings were our two preconditions. Our experiments and calculations were in tune with earlier results, while also supplemented them. If there was no fluid medium present during the experiments, the rotations or impacts with large kinetic energy revealed new ways of aggregation. In this way small and middle scale closed shapes could form, and not only ring shapes occurred, but some higher energy states, like droplet or D letter forms as well, which were stabilised by potential barriers and the mechanical interactions between the balls.

There are many studies concerning the stability and behaviour of the clusters of macroscopic magnetic dipoles (balls or cylinders), [26–28]. The method outlined here can support the theory by the macroscopic realization and direct observation of the aggregation process.

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References

- [1] George M W *et al* 2002 Self assembly at all scales *Science* **295** 2418
- [2] Zhang S 2003 Fabrication of novel biomaterials through molecular self-assembly *Nat. Biotechnol.* **21** 1171–8
- [3] Ku J Y, Aruguete D M, Alivisatos A P and Geissler P L 2011 Self-assembly of magnetic nanoparticles in evaporating solution *J. Am. Chem. Soc.* **133** 838–48
- [4] Randall M, Hui S S, Samanta B, Rotello V M and Yellen B B 2009 Magnetic assembly of colloidal superstructures with multipole symmetry *Nature* **457** 999–1002
- [5] Dominik C and Nübold H 2002 Magnetic aggregation I: aggregation dynamics and numerical modelling *Icarus* **157** 1173–86
- [6] Philippe A P and Maas Magnetic D 2002 Colloids from magnetotactic bacteria: chain formation and colloidal stability 9978 *Langmuir* **18** 9977–84
- [7] Timonen J V I, Latikka M, Leibler L, Ras R H A and Ikkala O 2013 Switchable static and dynamic self-assembly of magnetic droplets on superhydrophobic surfaces *Science* **341** 253–7
- [8] Ganguly R, Zellmer B and Puri I K 2005 Field-induced self-assembled ferrofluid aggregation in pulsatile flow *Citation: Physics of Fluids* **17** 097104
- [9] Elkady A S, Kakova L and Zubarev A 2015 On the self-assembly of net-like nanostructures in ferrofluids *Physica A: Statistical Mechanics and Its Applications* **428** 257–65
- [10] Wollny K, Läger J and Huck S 2002 Magneto sweep – a new method for characterizing the viscoelastic properties of magneto-rheological fluids *Appl. Rheol* **12** 25–31
- [11] Tripp S L, Pusztay S V, Ribbe A E and Wei A 2002 Self-assembly of cobalt nanoparticle rings *J. Am. Chem. Soc.* **124** 7914–5
- [12] Tripp S L, Dunin-Borkowski R E and Wei A 2003 Flux closure in self-assembled cobalt nanoparticle rings *Angew. Chem. Int. Ed.* **42** 5591–3
- [13] Jinhao G, Bei Z, Xixiang Z and Bing X Prof 2006 Agnetic-dipolar-interaction-induced self-assembly affords wires of hollow nanocrystals of cobalt selenide *Angew. Chem.* **45** 1220–3
- [14] Weijia Wen F, Kun K F, Pál D W, Zheng and Tu K N 1999 Aggregation kinetics and stability of structures formed by magnetic microspheres *Phys. Rev. E* **59** R4758–61
- [15] Pál G, Kun F, Varga I, Sohler D and Sun G 2011 Attraction-driven aggregation of dipolar particles in an external magnetic field *Phys. Rev. E* **83** 061504

- [16] Varga I, Yamada H, Kun F, Matuttis H-G and Ito N 2005 Structure formation in a binary monolayer of dipolar particles *Phys. Rev. E* **71** 051405
- [17] Varga I, Yoshioka N, Kun F, Gang S and Ito N 2007 Structure and kinetics of heteroaggregation in binary dipolar monolayers *J. Stat. Mech.* P09015
- [18] Morimoto H and Maekawa T 2000 Cluster structures and cluster–cluster aggregations in a two-dimensional ferromagnetic colloidal system *J. Phys. A: Math. Gen.* **33** 247–58
- [19] Ghazali A and Levy J-C 2003 Two-dimensional arrangements of magnetic nanoparticles *Phys. Rev. B* **67** 064409
- [20] Edwards B F, Riffe D M, Ji J-Y and William A 2017 Booth Interactions between uniformly magnetized spheres *Am. J. Phys.* **85** 130
- [21] Messina R and Khalil L A 2014 Self-assembly of magnetic balls: from chains to tubes *Phys. Rev. E* **89** 011202
- [22] Friedrich T, Rehberg I and Richter R 2015 Comment on ‘Self-assembly of magnetic balls: from chains to tubes’ *Phys. Rev. E* **91** 057201
- [23] Messina R and Stanković I 2015 Reply to ‘Comment on ‘Self-assembly of magnetic balls: From chains to tubes’’ *Phys. Rev. E* **91** 057202
- [24] Messina R and Stanković I 2015 Self-assembly of magnetic spheres in two dimensions: the relevance of onion-like structures *EPL* **110** 4
- [25] Schönke J, Schneider T M and Rehberg I 2015 Infinite geometric frustration in a cubic dipole cluster *Phys. Rev.* **91** 020410
- [26] Schönke J and Fried E 2017 Stability of vertical magnetic chains *Proc. R. Soc. A* **473** 20160703
- [27] Vella D, du Pontavice E, Hall C L and Goriely A 2014 The magneto-elastica: from self-buckling to self-assembly *Proc. R. Soc. A* **470** 20130609
- [28] Taberlet N, Ferrand J and Plihon N 2018 Stability analysis of an array of magnets: when will it jump? *Phys. Rev. Lett.* **120** 264301