A Tuneable Magnetic Domain Wall Conduit
Regulating Nanoparticle Diffusion

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Abstract

We demonstrate a general and robust method to confine on a plane strongly diffusing submicrometer particles in water by using size tunable magnetic channels. These virtual conduits are realized with pairs of movable Bloch walls (BW) located within an epitaxially grown ferrite garnet film. We show that, once inside the magnetic conduit, the particles experience an effective local parabolic potential in the transverse direction, while freely diffusing along the conduit. The stiffness of the magnetic potential is determined as a function of field amplitude which varies the width of the magnetic...
channel, and precise control of the degree of confinement is demonstrated by tuning the applied field. The magnetic conduit is then used to realize single files of non-passing particles and to induce periodic condensation of an ensemble of particles into parallel stripes in a completely controllable and reversible manner.

**Keywords**

Diffusion, Magnetic thin films, Domain Walls, nanofluidics

The ability to confine nanoscale particles in a fluid medium is of critical importance in a number of fields related to microfluidics, optics and biophysics. Optical tweezers based on focusing an intense laser beam to a diffraction limited spot, demonstrated in the past trapping, rotation, and transport of microscopic particles in water. Confinement of multiple microspheres in two and three dimensions has also been obtained with more sophisticated techniques such as fast scanning beams and holographic optical tweezers. However, reducing the size of the colloidal particles to the nanoscale makes stable trapping via optical means difficult, due to both the increase of thermal fluctuations and the need for large and localized optical force gradients. While individual gold and silver nanoparticles have been immobilized, confinement of the particles over an extended area, much larger than a single trapping spot, remains elusive.

Recent experiments using nanostructured slits made by electron-beam lithography, propose a different, non optical, approach to levitate and confine single nanoparticles along predefined tracks. These topographic reliefs induce a spatial modulation of the electrostatic potential, which attracts charged nanoparticles located at a close distance. However, the dynamics of colloidal particles moving near lithographic structures can be affected by the presence of the hard-walls due to hydrodynamic or steric interactions, and be rather sensitive to the ionic concentration of the dispersing medium.

Magnetic manipulation of polarizable particles is an alternative approach to control colloidal matter not relying on the particle surface charge or the dielectric constant, but rather on its
magnetic content. Magnetic nanoparticles can find applications in disparate fields, from spintronics,\textsuperscript{17} to biomedical research,\textsuperscript{18} hyperthermia,\textsuperscript{19} or as active propellers.\textsuperscript{20} While several approaches demonstrated forms of controlled trapping of microscale colloids above magnetic tracks,\textsuperscript{21–26} to our knowledge, only Ommering and coworkers\textsuperscript{27} were able to confine a single magnetic nanoparticle on a microfluidic chip.

Here we demonstrate a different approach which allows to obtain stable confinement in a fluidic environment of magnetic particles on a plane. Our technique is based on the use of remotely controllable domain walls in a ferromagnetic structured substrate, namely a uniaxial ferrite garnet film (FGF). The research on domain wall dynamics in magnetic systems has recently drawn much interest because of new applications in spintronics,\textsuperscript{28} logic devices,\textsuperscript{29,30} nanowires\textsuperscript{31,32} and ultracold atoms.\textsuperscript{33} Our magnetic conduit makes use of Bloch walls (BWs), i.e. transition regions where the magnetization rotates by 180 degrees through the wall separating opposite magnetized domains. The BWs in the used FGF can be displaced by applying relatively small external field, and they generate strong local stray fields capable to trap and confine magnetic microspheres.\textsuperscript{34} In this work we show that these highly functional features can be extended to nanoscale system, by tailoring the magnetic attraction of the film. In particular, we create a magnetic conduit for submicrometer particle motion using parallel planar BWs which provide locally a tuneable parabolic-like potential. These conduits are shown to be able to regulate the diffusive motion of an ensemble of particles, realizing e.g., single-file conduits for particle motion, where mutual passage is excluded. By applying an oscillating magnetic field, we vary the energy landscape and demonstrate periodic particle assembly and condensation into parallel bands.

The magnetic conduits are realized on the free surface of a FGF of composition $Y_{2.5}Bi_{0.5}Fe_{5-q}Ga_qO_{12}$ ($q = 0.5 – 1$), thickness $\sim 4\mu m$ and having a saturation magnetization (the FGF) of $M_s = 2.7\text{kA/m}$. The FGF is grown by dipping liquid phase epitaxy on a (111) oriented single crystal gadolinium gallium garnet (Gd$_3$Ga$_5$O$_{12}$) substrate.\textsuperscript{35} Fig.1(a) shows a typical transparent FGF sample used during the experiments. Due to the polar Faraday effect, the
magnetic domains in the FGF can be visualized via polarized light microscopy. As shown in Fig.1(b), under zero applied field the FGF is characterized by up- and down-magnetized domains which appear as stripes of black and white colors, repeating with a spatial periodicity $\lambda = 6.8 \mu m$. The BWs on the other hand, given their small width ($\sim 20 nm$), cannot be resolved by optical microscopy. The size of the magnetized domains can be easily manipulated via an external field. A magnetic field perpendicular to the film $H_z$, increases the width of the domains with magnetization parallel to the field ($\lambda_+$) and decreases the width of the antiparallel ones ($\lambda_-$). As shown in Fig.1(c), for $H_z < 3$ kA/m both, $\lambda_+$ and $\lambda_-$ vary linearly with $H_z$, and the total width $\lambda = \lambda_+ + \lambda_-$, remains essentially constant. In contrast, for higher field $\lambda_+$ and $\lambda$ diverge, showing large and irreversible deformations of the pattern. Beyond a critical value, the film becomes one single magnetized domain.85

We use three types of superparamagnetic polystyrene particles with diameters $d_p = 540nm$, 360nm and 270nm, COOH surface groups and $\sim 40\%$ wt. iron oxide content (Microparticles GmbH). Each type of particle is diluted in high deionized water and deposited above the FGF, where they sediment due to the magnetic attraction of the BWs. Once at the surface of the FGF, and in absence of any coating of the film, the strong attraction of the BWs rapidly immobilize the submicrometer particles, which show weak thermal fluctuations, see MovieS1 in the Supporting Information. In order to decrease the strong magnetic attraction, we coat the FGF film with a $h = 1.2 \mu m$ thick layer of a photoresist (AZ-1512 Microchem, Newton, MA), i.e. a light curable polymer matrix, using spin coating at 3000 rpm for 30s (Spinner Ws-650Sz, Laurell) and 5s of UV photo-crosslink (Mask Aligner MJB4, SUSS Microtec). Since the stray field of the FGF decreases exponentially from the surface, after coating the FGF with the polymer layer, the submicrometer particles display a stronger diffusive dynamics, but still remain two dimensionally (2d) confined above the FGF with negligible out of plane motion. Increasing the particle elevation from the film not only reduces the attractive force toward the substrate, but also modifies the shape of the potential, as schematically illustrated in Fig.1(d). Under a perpendicular field $H_z$, the magnetic potential at an elevation
\[ z_1 = \frac{d_p}{2} \] has minima located at the BWs and maxima between them. In contrast, at an elevation \( z_2 = \frac{d_p}{2} + h \), the potential minima (maxima) are now located at the center of the large (small) domains, and feature a local parabolic-like shape.

We can quantify the magnetostatic potential of the FGF, by calculating the energy of interaction \( U_m \) of one paramagnetic particle with the magnetic potential at a given elevation \( z \).

When subjected to a total magnetic field \( H_{\text{tot}} \), a paramagnetic nanoparticle acquires a dipole moment \( \mathbf{m} = V\chi H_{\text{tot}} \), pointing along the field direction. Here \( V = \pi d_p^3 / 6 \) is the particle volume, \( \chi \sim 2 \) the effective volume susceptibility\(^{27} \) and \( H_{\text{tot}} = H_a + H_s \) the sum of the applied field \( H_a \) and the stray field \( H_s \) of the FGF\(^{37} \). The interaction energy of the induced moment is thus given by, \[ U_m = -\frac{1}{2} \mu_w \mathbf{m} \cdot H_{\text{tot}} \] where \( \mu_w \sim \mu_0 = 4\pi \cdot 10^{-7} \text{H/m} \) is the magnetic permeability of the medium (water). Figs.2(a,c) show the normalized energy landscape \( U_m/k_B T \) numerically evaluated for a 360nm particle (elevation \( z = h + \frac{d_p}{2} = 0.20\lambda \)) in absence of an external field, Fig.2(a), and for an applied field of amplitude \( H_z = 620 \text{A/m} \), Fig.2(c). Here \( k_B \) is the Boltzmann constant and \( T = 293.15\text{K} \) the experimental temperature. In absence of the applied field, the FGF surface displays a sinusoidal-like potential at the particle elevation, blue curve in Fig.2(a), with several small wells which can be easily crossed by the fluctuating particle. Effectively, the corresponding particle trajectory in Fig.2(b) shows the submicrometer particle performing simple Brownian diffusion and passing the BWs without any perturbation of its random motion. In contrast, a static field \( H_z = 620 \text{A/m} \) is able to confine the particle motion on a narrow region between two BWs, i.e. along one ferromagnetic domain, Fig.1(d). Particles located above the domain of opposite magnetization, are repelled from there since, the energy landscape features a local maximum, blue curve in Fig.1(c).

In order to characterize the diffusive properties along the magnetic conduit, we measure the mean squared displacement (MSD) of the colloidal particles, \[ \langle \Delta y^2 \rangle = \langle [y(t) - y(0)]^2 \rangle \sim t^\alpha, \] being \( y \) the direction perpendicular to the magnetic conduit. Here \( \langle ... \rangle \) denotes an average over \( \sim 50 \) independent trajectories, and \( \alpha \) is the exponent of the power law which is used to
distinguish between normal diffusive ($\alpha = 1$) dynamics from the anomalous ones ($\alpha \neq 1$). In the first case, when $\langle \Delta y^2 \rangle \sim t$, one can extract the effective diffusion coefficient from the slope of the MSD as $D_y = \lim_{t \to \infty} \langle \Delta y^2 \rangle / 2t$. Fig. 3(a) shows several MSDs for a 360 nm particle in a magnetic conduit at different values of the applied field $H_z$. For $H_z = 0$, the motion is isotropic and the colloidal particles show the same diffusion coefficient in the 2d plane, $D_x = D_y = 0.78 \mu m^2/s$ for the 360 nm particle.

We characterize the zero field dynamics for all three types of paramagnetic colloids, and provide in Table 1 the comparison between the effective diffusion coefficients measured on top of a non-magnetic glass substrate and on top of the FGF. The first ones (column 2, Table 1) are closer to the theoretical predictions obtained from the Stokes-Einstein relationship given by $D_x = k_B T / \zeta$. Here $\zeta = 3\pi \eta f d_p$ is the friction coefficient of a particle immersed in water ($\eta = 10^{-3} Pa \cdot s$), and $f$ a correction factor which takes into account the proximity of the wall. For the glass substrate (column 3, Table 1) we take $f = 1$ since the particles easily detach from the plane in absence of any magnetic attraction. In contrast the FGF attracts the magnetic particles reducing their effective Brownian diffusion. Here the particles will experience higher friction since $f > 1$, and from the expression of $f$, we can estimate an average elevation of 80 nm of the nanoparticle surface from polymer film coating the FGF. The particle levitation above the FGF results from the balance between the magnetic attraction and the repulsion arising from electrostatic and steric interactions with the polymer film coating the FGF.

When applying the field, the particle motion is confined within the magnetic conduit, and the MSD rapidly saturates to a plateau which indicates the average maximum space the particle can explore. Assuming that the complex trapping potential can be approximated locally by a parabolic one of type $U_m(y) \sim \frac{1}{2} k_e y^2$, being $k_e$ the elastic constant, one can derive the corresponding MSD from the overdamped Langevin equation. For a single magnetic
particle in this potential one has two independent equations of motion,

\[ \zeta \frac{dx}{dt} = f_x(t) \]  
\[ \zeta \frac{dy}{dt} = -k_e y + f_y(t) , \]

(1)  
(2)

where \( f_i \) are the random forces \( (i = (x, y)) \) with properties, \( \langle f_i(t) \rangle = 0 \) and \( \langle f_i(t)f_i(t') \rangle = 2\zeta k_B T \delta(t - t') \). Solving these equations give, respectively

\[ \langle \Delta x^2 \rangle = 2 \frac{k_B T}{\zeta} t = 2Dt \]  
\[ \langle \Delta y^2 \rangle = \frac{k_B T}{k_e} \left[ 1 - \exp \left(-2k_e t/\zeta \right) \right] . \]

(3)  
(4)

We use Eq.(4) to fit all curves showed in Fig.3(a), by keeping constant \( \zeta = 5.2 \cdot 10^{-3} pN \cdot s/\mu m \), and varying \( k_e \) as a fitting parameter. The results are presented in Fig.3(b), which shows a linear relationship between \( k_e \) and the width of the magnetic conduit \( \lambda_+ \). As shown in the small inset in Fig.3(b), \( \lambda_+ \) which correspond to the sizes of the majority domains, varies linearly with the applied field \( H_z \). In order to resolve directly the parabolic shape of the potential, we also analyze the distribution of displacements when individual particles are trapped in the magnetic conduit. Fig.3(c) shows as a typical example the case for an applied field \( H_z = 620A/m \). The normalized potential was obtained by assuming that the probability density \( p(y) \), inset Fig.3(c), follows the Boltzmann distribution, \( p(y) = p(0) \exp \left(-U_m/k_B T \right) \). Inverting this equation gives the potential \( (U_m(y) - U_m(0))/k_B T = \log \left[p(0)/p(y) \right] , \) which is presented in Fig.3(c). Fitting this curve gives the value of \( k_e = 0.044pN/\mu m \), which is very close to the value obtained in the independent fit in Fig.3(b), \( k_e = 0.042pN/\mu m \) for \( H_z = 620A/m \). This result also enforces the hypothesis that that the complex magnetic potential can be approximated locally by a parabolic one.

We next explore the effect of increasing the particle concentration inside the magnetic conduit, for particle densities \( \rho < 0.6 \mu m^{-1} \), where \( \rho = N/L \) is the linear density and \( L \) the length
of the channel. We find that increasing the potential stiffness via the applied magnetic field, reduces the particle motion from 2d, where the submicrometer particles can pass each other, to strictly one dimension (1d), Fig.4(a). In the first case, two approaching particles can eventually exchange position, and this process occur by sliding laterally along the channel width rather than overriding one below the other. In the second case, the particles form a ”single file” system, i.e. they assemble into a 1d chain of diffusing submicrometer particles unable to pass each other. Single file diffusion is a phenomenon observed in a broad class of system, from zeolites,40 to ion channels,41 dipeptide,42,43 and carbon nanotubes.44 While the statistical properties of a single file of particles are well understood and experimentally demonstrated,45,46 the transition from single-file to 2d diffusion has been much less accessible,47 and was only recently realized with microscopic particles.48 Here we monitor this transition by measuring the particle distribution $P(y)$ along the channel width, Fig.4(b). In agreement with previous numerical results,47 we observe that as the confinement decreases, the distribution become broader, and its variance increases. Also, given the parabolic 1d confinement of the FGF, the distribution of particles $P(y)$ remain symmetric along the center of the magnetic conduit ($y = 0$). However, in contrast to the numerical results reported previously,47 beyond the crossover point from 2d to 1d, when the particles start to pass each other, we did not observe a bimodal distribution of the particle displacement. The particles did not form a two-chain structure which would cause two small peaks in the $P(y)$, as observed in simulation for an idealized system of repulsive microscopic colloids. This difference may arise from the fact that the inter-particle interactions are not strong but rather the system behaves as a confined hard sphere gas with a short repulsive potential resulting from steric interactions. We also shown that the particle distribution $P(x)$ along the channel, inset of Fig.4(b), behave as a Gaussian (continuous line), as predicted theoretically in the past.49,50 By analyzing the particle distribution in Fig.4(b) we find that the crossover from 2d to 1d occurs for field amplitude $H_z = 870 \text{A/m}$ which corresponds to a standard deviation $\sigma_x = 0.53 \mu m$ for the 360nm particles. Smaller particles 270nm require much stronger field
to be confined into a 1d line, $H_z > 1240\,\text{A/m}$, while larger ones ($540\,\text{nm}$) are already unable to pass each other for field $H_z > 530\,\text{A/m}$, inset Fig.4(c). This behaviour is a direct consequence of the linear dependence of the potential stiffness with the applied field, Fig.2(b), since smaller particles have larger thermal fluctuations and thus require an higher field to be confined into a 1d line. Moreover, the data in Fig.4(c) can be well fitted with an exponential curve, $H_z = H_0^z \exp(-d_p/\beta)$ which allow us to estimate as $d_p = 75\,\text{nm}$ the minimum particle size which would be possible to trap in a 1D single file for a maximum applied field of $H_z = 3\,\text{kA/m}$, beyond which the magnetic conduit start deforming.

For high concentration of particles, excluded volume effects become important and cause clogging inside the magnetic conduit, which causes freezing of the colloidal structure strongly reducing the particles diffusive motion. In Fig.5 we report the condensation of a large collection of $270\,\text{nm}$ sized particles on up- or down-magnetized domain depending on the direction of the applied field. Upon application of an oscillating field at an angular frequency $\omega = 2\pi\,\text{rad/s}$ and an amplitude $H_0^z = 1370\,\text{A/m}$, $H_z = H_0^z \sin(\omega t)$, the particles can be periodically translated from one domain to the next, since the magnetic energy landscape is inverted during each field period, changing maxima to minima and viceversa. As shown in VideoS4 in the Supporting Information, the particles perform a periodic motion between the magnetic domains, which is essentially synchronous with the driving field. Once inside a minima, the particles do not show any net motion and their dynamics is frozen within the cages created by the nearest particles. Thus stopping the field at a certain value allows to solidify the pattern in its current state, without loosing particles in the nearest domains. Since the BW response to the applied field can be considered instantaneous if compared to the particle self-diffusion time, this capability to condense particles pattern suggests new route of printing magnetic ink containing nanoparticles, or can be used for novel magneto-optical or biosensing applications.

In summary, we report a simple and robust method to confine strongly fluctuating magnetic submicrometer particles on a plane and tuning their diffusive motion from 1d to 2d. The
transition between these dimensions can be easily induced via application of a relatively small external field. Although our study is limited to particles which can be resolve by optical microscopy, separate model calculations (not presented here) suggest that also smaller particles can be equally confined in the magnetic conduits when adjusting their height above the FGF surface. Since the complex magnetic potential generated by pairs of BWs can be well approximated by an idealized parabolic well, the present pilot system could serve as a testbed for fundamental studies of transport and diffusion at the nanoscale. Finally, the implementation of different FGF film with in plane magnetization would in principle allow manipulation of a single particle within an ensemble using magnetic domain wall tips.\cite{51}

Table 1: Effective translational diffusion coefficients at $H_z = 0$ above a glass plate ($D_g$), and above the FGF ($D_{FGF}$). The theoretical values are calculated for $f = 1$ ($D^{th}_g$) and for a particle elevation of 80nm ($D^{th}_{FGF}$).

| Particle size (nm) | $D_g$ ($\mu$m$^2$/s) | $D^{th}_g$ ($\mu$m$^2$/s) | $D_{FGF}$ ($\mu$m$^2$/s) | $D^{th}_{FGF}$ ($\mu$m$^2$/s) |
|-------------------|---------------------|--------------------------|--------------------------|--------------------------|
| 270               | 1.58 ± 0.05         | 1.61                     | 1.04 ± 0.05              | 1.03                     |
| 360               | 1.18 ± 0.03         | 1.20                     | 0.78 ± 0.03              | 0.72                     |
| 540               | 0.81 ± 0.02         | 0.80                     | 0.52 ± 0.03              | 0.44                     |

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Supporting Information Available

The following files are available free of charge. Four experimental videos (.WMF) showing the dynamics of magnetic colloidal particles above the FGF film.
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Figure 1: (a) Image of a ferrite garnet film (FGF) on a gadolinium gallium garnet substrate, scale bar is 1 cm. (b) Polarization microscope image showing a 57 × 47 µm² section of a magnetic stripe pattern in a FGF film, scale bar is λ = 6.8 µm. (c) Domain width versus amplitude of the magnetic field applied perpendicular to the film, \( H_z \). The applied field increases (decreases) the width of the domain with parallel (antiparallel) magnetization, \( \lambda_+ \) (\( \lambda_- \), respectively), being \( \lambda = \lambda_+ + \lambda_- \). \( H_c = 13900 \text{ A/m} \) denotes the critical field at which all the antiparallel (grey) domains disappear. Shaded green area indicates the region where the measurements are taken. (d) Schematic of a FGF film with one magnetic particle and subjected to the perpendicular field \( H_z \). Red arrows within the FGF denote the magnetization direction, dashed and continuous blue lines refer to the magnetic potential \( U_m \) for a submicrometer particle at the FGF surface, elevation \( z_1 \), and above the photoresist layer at elevation \( z_2 = z_1 + h \), with \( h = 1.2 \mu m \).
Figure 2: (a,c) Color coded energy landscape $U_m/k_B T$ of one submicrometer particle in absence of field, $H_z = 0$ (a), and under an applied field $H_z = 620 \text{A/m}$ (c). Continuous blue lines indicate the corresponding scaled potential $U_m - \langle U_m \rangle$ plotted at the particle elevation, $z = 0.20 \lambda$. (b,d) Corresponding microscope snapshots of one 360nm particle with superimposed the trajectory (green line) for $H_z = 0$ (b), and $H_z = 620 \text{A/m}$ (d). Scale bar is 5 $\mu$m, dashed lines denote the location of the BWs. See also corresponding MovieS2 in the Supporting Information.
Figure 3: (a) Log-log plot of the mean square displacement (MSD$_y$) for different applied magnetic fields. Continuous lines are fit to the data using Eq.(4) in the main text. In absence of field (black squares) the particle displays normal diffusion with slope $\alpha = 1$. (b) Spring constant $k_e$ measured at different width $\lambda_+$ of the magnetic channel for a 360nm particle. Continuous red line denotes linear fit to the data. Inset shows the linear relationship between $\lambda_+$ and the applied field $H_z$ for the range of field amplitude used. (c) Normalized magnetic potential of a 360nm particle measured from the particle fluctuations. The red line represents a parabolic fit to the data using a stiffness $k_e = 0.044$ pN/µm for an applied field $H_z = 620$ A/m. Small inset at the top shows the normalized position distribution, fitted to a Gaussian function (green line) having dispersion $\sigma_y = 0.36$ µm.
Figure 4: (a) Snapshot of a single file composed by 17 particles (360 nm) subjected to a field $H_z = 1240$ A/m. Scale bar is 5 µm. Corresponding VideoS3 is included in the Supporting Information. (b) Probability distribution of the particle position $P(y)$ along the direction perpendicular to the magnetic conduit for a linear density $\rho \sim 0.3$ µm$^{-1}$. Inset shows the probability distribution $P(x)$ along the magnetic channel for an applied field $H_z = 2600$ A/m, with a Gaussian fit (continuous line). (c) Field dependence of the transition region from 2d to 1d motion versus particle size $d_p$. The continuous red line is an exponential fit $H_z = H^0_z \exp\left(-d_p/\beta\right)$ to the data points, with $H^0_z = 3158$ A/m and $\beta = 285$ nm.
Figure 5: Top: series of microscope images of a small section (16.8 × 19.2 µm²) of an ensemble of 270 nm particles deposited above a FGF film at different values of the applied field. See VideoS4 in the Supporting Information. Bottom: external magnetic field $H_z$ oscillating around the z direction with angular frequency $\omega = 2\pi \text{rad/s}$ and amplitude $H_z^0 = 1370 \text{A/m}$. Small inset shows the full overview (64.3 × 48.2 µm²) of the condensed pattern at $t = 0.25s$, scale bar is equal to $\lambda$. Corresponding VideoS4 is included in the Supporting Information.