Magnetophoretic lensing by concentric topographic cylinders of perpendicular magnetic anisotropy multilayers

Maciej Urbaniak,^{1,a)} Dennis Holzinger,² Arno Ehresmann,²
 and Feliks Stobiecki¹

¹*Institute of Molecular Physics, Polish Academy of Sciences, ul. M. Smoluchowskiego 17,
 60-179 Poznań, Poland*

²*Department of Physics and Center for Interdisciplinary Nanostructure Science
 and Technology (CINaT), University of Kassel, Heinrich-Plett-Str. 40,
 D-34132 Kassel, Germany*

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Colloidal magnetophoretic lensing of water suspended micrometer-sized superparamagnetic beads (SPBs) above a topographically patterned magnetic thin film system with perpendicular magnetic anisotropy is demonstrated. The magnetic pattern consisting of concentric annuli of micron-sized widths has been superimposed with a rotating external magnetic field, and it is shown that the trajectories of the SPBs above this structure are similar to light rays in an optical focusing lens. SPB trajectories converge towards the central region and have divergent trajectories while passing the center. The experimental findings are corroborated by a quantitative model for the SPB trajectories. The magnetophoretic lensing effect leads to a high SPB concentration in the center of the pattern and may be useful for applications where SPBs have to approach each other in a controlled way. *Published by AIP Publishing.*
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I. INTRODUCTION

Monitoring and therapy control of diseases are among the most important health policy issues. Reliable point-of-care diagnostic tools are needed to help to secure broad access to appropriate medical treatment.¹ Possible solutions for cost effective sample analysis are lab-on-a-chip devices employing microfluidic sample (e.g., blood serum and urine) processing combined with portable communication devices.² For many years, devices employing magnetophoresis, i.e., remotely controllable transport of magnetic particles using either homogeneous or inhomogeneous external magnetic fields, have been intensively investigated.^{3–8} Experiments most often use micron-sized biocompatible superparamagnetic beads (SPBs) functionalized^{9,10} with capture molecules to bind analytes. Besides simply using external magnetic gradient fields, like in magnetic targeting of anticancer agents to malignant tissue,¹¹ a more advanced way to move SPBs makes use of a superposition of substrate based local but static magnetic field sources^{12–14} (magnetic stray field landscapes, MFLs) and temporally varying homogeneous external magnetic fields.¹⁵ The static MFLs provided by various geometries of either topographically shaped micromagnets or artificial magnetic domain patterns possess comparably large field gradients due to small length scales, leading to significantly higher bead velocities when compared to external field gradients. The AC external field shifts the MFL across the substrate, resulting in a dynamic switching of the beads' magnetic potential energy landscape, therefore inducing a controlled and directed movement in close proximity to the substrate plane.

In one detection scheme for analyte molecules with two binding sites,¹⁶ one SPB functionalized with a capture molecule fitting to one analyte binding site may be bound to a second SPB with a capture molecule fitting to a second binding site via an analyte molecule bridge.¹⁷

^{a)}Electronic mail: urbaniak@ifmpan.poznan.pl. URL: <http://www.ifmpan.poznan.pl/~urbaniak>

Such a SPB dimer can be easily detected by optical microscopy, or it can be separated from single SPB using its deviating magnetophoretic mobility.¹⁸ A critical requirement for such an assay is a high probability for physical proximity between the two types of SPBs for high interaction rates. For enhancing the interaction rate, it has recently been suggested to use two types of SPBs with differing magnetic properties.¹⁶

Here, an alternative approach is presented in which SPBs with the same magnetic characteristics can be used. To increase the probability that SPBs approach each other in a certain substrate area, a topographic pattern of concentrically arranged hollow cylinders has been engineered to produce a MFL, leading to converging SPB trajectories into a certain substrate area and to diverging trajectories when this area has been passed by the SPBs. As a result, the movement of SPBs above this structure when superimposed with rotational external magnetic fields is similar to the path of rays in an optical focusing lens. Hence, SPBs are concentrated within the central area of the cylinder pattern.

In the present work, Au/Co/Au multilayers (MLs) with perpendicular magnetic anisotropy (PMA)¹⁹ have been topographically patterned into an array of concentric annular cylinders (referred to as cylinders hereafter for simplicity). We will show that this cylindrical arrangement of micromagnets characterized by a C_∞ symmetry axis leads to spatial focusing of the SPB trajectories analogous to an optical lens. In this respect, the overall SPB movement above the here suggested structure can be classified into three parts: spatial convergence, external gradient field magnetophoresis, and interaction between SPBs within the focus region and spatial divergence after having passed the focus region. The observed SPB trajectories will be quantitatively modeled by taking into account a spatial and temporal change of all relevant forces acting on the SPBs.

II. EXPERIMENTAL

The details of the experimental procedure are described in Ref. 19. Here, we only give a brief summary. Figure 1 schematically shows the geometry of the experiment. The thin film Ti(4 nm)/Au(60 nm)/[Co(0.7 nm)/Au(1 nm)] $\times$ 3 system with perpendicular magnetic anisotropy has been deposited on $10 \times 10 \text{ mm}^2$ Si(100) substrates covered with a patterned PMMA electron resist using DC magnetron sputtering with deposition rates of 0.08 nm s^{-1} for Ti, 0.11 nm s^{-1} for Au, and 0.09 nm s^{-1} for Co, respectively. The preceding lithography was carried out using a FEI Nova NanoSEM 650 electron microscope equipped with a Raith ELPHY Plus lithography system. After sputter deposition of the MLs, a lift-off process created sets of concentric hollow cylinders with several micrometer widths, usually slightly smaller spacing between the edges of neighboring cylinders [see Fig. 2 (Multimedia view)], and a total height

FIG. 1. Sketch of the model used to theoretically describe the SPB movement. The concentric thin hollow cylinders of width w and equal spacing in-between are magnetized parallelly to the z -axis. The external magnetic field is applied parallel to the xz -plane. The SPBs move under the action of the external field, due to the temporal changes of their magnetic potential energy landscape. The field direction changes in a step-like manner: $(+H_x, +H_z) \rightarrow (+H_x, -H_z) \rightarrow (-H_x, -H_z) \rightarrow (-H_x, +H_z)$. The two black lines illustrate the exemplary simulated trajectories of the $4 \mu\text{m}$ sized beads (moving away from the center of cylinders) placed initially at $r = 43 \mu\text{m}$ and $z = 2.4 \mu\text{m}$: for $\phi = 0^\circ$ and $\phi = 60^\circ$. Both the lines correspond to the data shown in Fig. 8. Note that the thickness of the cylinders is exaggerated in the image.

FIG. 2. Last frame from a movie showing the movement of the $4\ \mu\text{m}$ diameter SPBs, which are immersed in water, over the Au/Co/Au ML topographically patterned into a set of concentric hollow cylinders (see the movie in the [supplementary material](#)). The cylinders are $7\ \mu\text{m}$ wide, and the spacing between edges of the neighboring cylinders is $2\ \mu\text{m}$. The external field ($H_x = \pm 1.2\ \text{kA/m}$ and $H_z = \pm 1.6\ \text{kA/m}$) changed its direction clockwise, looking from the $-y$ direction, starting from a $(++)$ orientation. The yellow, red, and green lines show the in-plane projections of automatically tracked experimental trajectories of exemplary beads (see Sec. II). Note that all the beads move from left to right. The blue squares indicate the end positions of the tracked SPBs, which do not necessarily correspond to the final bead position in the movie in case that the tracker lost the bead position during the tracking routine. The cylindrical coordinate system (r, ϕ, z) used in the analysis is also outlined. Multimedia view: <https://doi.org/10.1063/1.5034516.1>

above the substrate plane of $69\ \text{nm}$. For the magnetophoresis experiments, 1 and $4\ \mu\text{m}$ sized core-shell⁹ beads from chemicell²⁰ and micromod²¹ were used, respectively. In the case of the $1\ \mu\text{m}$ beads, the beads' magnetic content is located within the inner core region of the bead, whereas for the $4\ \mu\text{m}$ sized SPBs, a non-magnetic core is surrounded by a $100\ \text{nm}$ thick spherical shell, in which superparamagnetic iron oxide nanoparticles are dispersed.¹⁹ For the experiments, the SPBs were suspended in deionized water. The optical microscope used for the particle transport observation ($40\times$ optical magnification) was equipped with a set of coils to generate the AC external magnetic field sequence. The coil pairs independently produced varying external magnetic fields perpendicular to the sample's surface (z -coordinate) and a field component in a direction coplanar with the substrate surface (x -coordinate, see Fig. 1). During the experiment, periodic square wave current pulses, with rise and fall times below $0.5\ \text{ms}$,¹² were applied to both the coils, with a phase-shift between them of a quarter period, resulting in the following sequence: $(+H_x, +H_z) \rightarrow (+H_x, -H_z) \rightarrow (-H_x, -H_z) \rightarrow (-H_x, +H_z)$. This sequence created a stepwise rotating magnetic field. The magnetic field strengths [see the caption of Fig. 2 (Multimedia view)] were chosen as to not influence the magnetization state of the stripes possessing a switching field of $H_S \approx 40\ \text{kA/m}$, as determined by perpendicular magneto-optical Kerr-effect (P-MOKE) magnetometry. As we showed earlier, the patterning of such MLs does not significantly influence the MLs' magnetic properties.²² Videos of the particle transport were recorded using an Optronis CR450x2 camera with a $1000\ \text{fps}$ frame rate, and each record lasted approx. $9\ \text{s}$. In-plane (xy) trajectories of the SPBs were automatically tracked, whenever possible, using the software Video Spot Tracker²³ from NIH. We used a symmetric kernel with "follow jumps" feature enabled. Since the initial position of SPBs relative to the patterned cylinders could not be reproducibly set in the experiments, we applied the following procedure to estimate a maximal uncertainty for the observed SPB trajectories, where three trajectories for SPBs travelling at $\phi \approx 212^\circ$ [see Fig. 2 (Multimedia view), yellow, red, and green trajectories] were chosen. Neglecting that the MFL is not exactly periodic, we assumed that all three SPBs should move in almost the same manner when driven by the external field. When shifted to the common starting point, the three trajectories $\vec{r}_i(t)$ ($i = 1, 2, 3$) differ by not more than $6\ \mu\text{m}$ (for $t \in [0, 4\ \text{s}]$: $|r_i(t) - r_j(t)| < 6\ \mu\text{m}$; $i, j = 1, 2, 3$; $i \neq j$).

Consequently, the maximum uncertainty of the tracked SPB position at any time was estimated to be $3\ \mu\text{m}$. Note that the maximum uncertainty already takes into account deviations in the SPB trajectory due to diffusion (at maximum $1\ \mu\text{m}$ for a $4\ \mu\text{m}$ sized SPB within 5 s), variations in the SPB size and the magnetic content, interaction between neighboring SPBs, and tracking issues due to low contrast between SPBs and the magnetic substrate. Based on this, the error of the average velocity of the SPBs was derived from the estimated uncertainty in the SPB trajectory (Fig. 4).

III. CALCULATIONS

Modeling the dynamic transport of SPBs floating in a liquid (here: water) over a micrometer-sized magnetic pattern requires the calculation of the magnetic stray field landscape produced by the pattern. As the micromagnetic pattern is cylindrically symmetric, the emerging MFL is cylindrically symmetric as well. This is, however, not the case for the superimposed external field, which rotates in a stepwise manner in the xz -plane. If the lateral coordinate system of Fig. 2 (Multimedia view) is used, it is possible to reduce the problem of the magnetic field or the magnetic potential energy landscape of the SPBs to a one dimensional representation for distinct angles ϕ . To compute the radial magnetic fields of the axially magnetized hollow cylinders, we used Eq. (4) from Ref. 24 (Appendix A), and to calculate the axial field components, we employed Eq. (16) from Ref. 25 (Appendix B, Ref. 26). The equations were derived under the assumption of a uniformly magnetized magnetic pattern with perpendicular to plane magnetization. In the simulation, the cylinders have a thickness equal to the total thickness of the three Co layers. Five cylinders were included in the calculations to demonstrate the main features of the SPB transport. However, when a comparison with the experiment was made (see Figs. 9 and 10) and to avoid edge effects, the number of cylinders was increased so that the outermost point of the experimental trajectory was at least $15\ \mu\text{m}$ away from the outer edge of the largest cylinder. The magnetic force acting on the beads was calculated by the negative gradient of the SPB's magnetic potential energy²⁷ (see Figs. 5 and 6),

$$\vec{F}_m = \frac{1}{2\mu_0} \chi V \nabla(\vec{B}^2), \quad (1)$$

where χ is the effective magnetic susceptibility (this quantity corresponds in our case to the difference in the bead susceptibility and the fluid susceptibility; for water, $\chi \approx -9 \times 10^{-6}$ —Ref. 28) which we assumed to be constant for weak magnetic fields acting on the beads in our experiment, and V is the bead's magnetic volume (compare Refs. 29 and 30). In the simulations, to prove the applicability of the design for magnetophoresis, we used SPB with a diameter of $4\ \mu\text{m}$. The results for SPBs with a diameter of $1\ \mu\text{m}$ are shown in the [supplementary material](#); they prove that the character of trajectories is not size-dependent. In the present calculation, the SPB is approximated by a magnetic point dipole located in the bead center which approximates well the SPBs of core-shell type with relatively small magnetic core diameters. A comparative calculation for a model bead divided into 16 cells showed no change in the essential findings of the present work (see Fig. 1 of the [supplementary material](#))

In the simulations, magnetic forces exerted by the magnetized cylinders, viscous forces due to the aqueous SPB environment, electrostatic forces (double-layer repulsion) between surfaces of the beads and the substrate,³¹ as well as buoyancy, and gravity were taken into account. The SPB density typically ranges between $1.1 \times 10^3\ \text{kg m}^{-3}$ (Ref. 12) and $2.25 \times 10^3\ \text{kg m}^{-3}$ (Ref. 20) for polystyrene and silica based beads, respectively. For the calculations, we therefore assumed an intermediate density of $1.7 \times 10^3\ \text{kg m}^{-3}$. The viscous force acting on spherical beads of radius r_B moving with velocity v in the aqueous environment of viscosity η was calculated from Stokes's law³²

$$\vec{F}_S = 6\pi r_B \eta f_R(z_B - r_B) \vec{v}, \quad (2)$$

where f_R is the drag coefficient which depends on the distance $z_B - r_B$ between the SPB surface and the substrate surface—see Eq. (6) in Ref. 32. The electrostatic force between the bead and the substrate, which in general can be either repulsive or attractive, was estimated using the Derjaguin approximation of the double layer repulsion [Eq. (10.18) and p. 243 in Ref. 31],

$$F_D = (128k_B T \rho_\infty \pi r_B \gamma_B \gamma_s / \kappa) e^{-\kappa(z_B - r_B)}, \quad (3)$$

where $\gamma = \tanh(\nu e \psi_0 / 4k_B T)$ is the surface energy [different for the bead (B) and for the substrate(s)], in which the surface potential ψ_0 has been approximated by the corresponding zeta-potential ζ , where $1/\kappa$ is the Debye length (300 nm in our simulation, but strongly pH dependent³¹), ν is the valency of the ions, ρ_∞ is the ionic concentration in the bulk liquid, and z_B is the distance of the bead's center to the substrate. To estimate the electrostatic force from Eq. (3), we take $\zeta_B = -32$ mV which corresponds to the SiMAG²⁰ beads, $\zeta_s = -9.2$ mV (Ref. 33) for an Au surface in deionized water (the Co/Au multilayer system was covered with Au) and $\zeta_s = -20$ mV for the silicon substrate³⁴ (corresponding to the zeta-potential of the sample areas not covered by the metallic multilayer system). Note that Eq. (3) was derived for an infinitely extended plane and is only an approximation of the situation observed in the experiment [Fig. 2 (Multimedia view)] where only about 78% of the substrate's active area is covered by the metallic multilayer. Assuming that the resultant zeta potential of the patterned structure is proportional to the relative areas of the Si and Au covered parts, the prefactor of the exponent in Eq. (3), in the case of the 4 μ m sized SPBs, is approx. 2.15×10^{-11} N. In the simulations, we used prefactor values ranging from 2.6×10^{-12} N to 2.6×10^{-11} N, and as the electrostatic interaction strongly depends on the chosen substrate-SPB-liquid system, Eq. (3) can be understood as an estimate for these repulsive forces (compare Fig. 2 in the [supplementary material](#)).

In the experiment, the external magnetic field modulates the energy landscape of the bead in a stepwise manner which affects the direction and strength of the magnetic forces acting on the SPB.^{12,13,19,35} Magnetic field switching occurs with typical switching times of less than 0.5 ms (Ref. 12) and periods of 0.25 s with a constant magnetic field landscape. As the switching time is considerably faster than any timescale relevant for the observed SPB motion, the switching time intervals will be excluded in the following discussion. In the periods of constant external field, the beads move along the trajectories determined by the magnetic stray field landscape generated by the substrate with a superimposed external field. To calculate the trajectory of a SPB, an obvious approach would be to numerically integrate the equation of motion, which includes the force described by Eq. (2) and which collects all other remaining forces into an effective force $\vec{F}_{ext}$ [Eqs. (1) and (3), together with gravity and buoyancy],

$$\vec{F}_S + \vec{F}_{ext} = \frac{4}{3} \pi r_B^3 \rho \cdot \ddot{\vec{r}}, \quad (4)$$

where ρ is the SPB density and $\vec{r}$ is the position of its center. Assuming that these forces do not depend on the SPB position, Eq. (4) can be solved analytically. This yields the following expression for the SPB velocity (assuming rest at $\vec{r} = 0$ m at $t = 0$ s):

$$\dot{\vec{r}}(t) = \frac{\vec{F}_{ext}}{6f_R \eta \pi r_B} \left[1 - \exp\left(-\frac{9f_R \eta}{2\rho r_B^3} \cdot t\right) \right] = \vec{v}_{te} (1 - e^{-t/\tau}), \quad (5)$$

where $\vec{v}_{te} = \vec{F}_{ext} / 6f_R \eta \pi r_B$ is the SPB steady-state velocity and τ is a measure for the corresponding acceleration time. After some 4.6 τ , the SPB attains 99% of v_{te} . For the SPB diameter and density used in the simulations, assuming $f_R = 1$, this time is 7 μ s, and the distance traveled during that time, taking external forces $\vec{F}_{ext}$ from the simulation ($\sim 2 \times 10^{-12}$ N), is less than $v_{te} \cdot 5\tau$, i.e., less than 1 nm which is negligible in comparison to the dimensions or even the widths of the patterned cylinders. Consequently, we assume that the bead movement can be described by the position dependent steady state-velocity v_{te} which can be calculated by the balance of forces described by $\vec{F}_S$ and $\vec{F}_{ext}$ [compare Eq. (4)]. Starting from an initial arbitrary xyz -position of the

bead, we calculate $\vec{v}_{te}(\vec{r}, t)$ and advance the bead's position with an adaptive algorithm:^{36,37} the time step Δt is adapted so that the difference in a spatial step along any of the Cartesian directions between a trajectory traveled during a time step Δt and two successive trajectories traveled during two consecutive time steps of $\frac{1}{2}\Delta t$ is less than $0.01 \mu\text{m}$. Usually, Δt is within 0.1–20 ms. Note that the explicit dependence of v_{te} on t reflects the periodic changes of the external field direction used to induce the magnetophoretic movement. In the simulation, we switch on the external field H_{ext} after allowing the SPB to approach its equilibrium position (reached, per definition, when none of the Cartesian components of its velocity exceeds $0.05 \mu\text{m s}^{-1}$), which does not typically coincide with the position in the presence of the external field. Note that only a single SPB is included in the simulations, and so, the interactions between beads are not considered. Finally, we obtain the three dimensional trajectory of the bead: $\vec{r} = \vec{r}(t)$ (Figs. 7–12).

IV. RESULTS AND DISCUSSION

The experimentally tracked trajectories of the SPBs shown in Fig. 2 (Multimedia view) indicate that the SPB current is locally perpendicular to the edges of the magnetic cylinders when an external magnetic field sequence is applied within the xz -plane. Such a behavior is also observed for magnetophoresis experiments above engineered parallel-stripe domains.^{13,19} The SPBs that are positioned on the left hand side of the structure are directed towards the center, while those to the right are continuously spread over the cylinder's perimeter (see the movie for $1 \mu\text{m}$ sized SPBs in the [supplementary material](#)). Note that some of the SPBs (the uppermost trajectory and the two trajectories below it) move along the edges of the cylinders, mainly in the region, where the edge of the structure is almost parallel to the plane of the external driving field, i.e., along the x -direction. Magnetic potential energy of the SPBs is degenerated for these positions, similar to a potential well of equal depth. Note that within these regions, e.g., thermal kinetics could further influence the SPB motion.¹¹ Figure 3, where the

FIG. 3. Image sequence extracted from a movie for the $4 \mu\text{m}$ sized SPBs [compare Fig. 2 (Multimedia view)] at equal time intervals of 125 ms recorded for a driving field of 1 Hz, i.e., for one field period, with field amplitudes of $H_x = 1.2 \text{ kA/m}$ and $H_z = 1.6 \text{ kA/m}$. The width of the images is $55 \mu\text{m}$.

SPB dynamics for a relatively low external field frequency of 1 Hz is shown, illustrates that within one period of the driving field, the SPBs travel a distance that is equal to one spatial period of the cylinder structure of the magnetic substrate. For such low frequencies f , the SPBs are able to follow the field changes even in case that they are located at an angle $\phi > 0^\circ$ with respect to the x -axis, i.e., the xz plane of the external magnetic field, and the resulting SPB trajectory is tilted with respect to the x -axis [Fig. 4(a)]. When using higher frequencies [Fig. 4(b)], the average velocity of the SPBs is increased up to a critical frequency, and in the case of the $4\ \mu\text{m}$ sized beads, it drops down to zero for $f = 100\ \text{Hz}$. For that frequency, almost all SPBs perform an oscillatory motion within one spatial period of the underlying magnetic substrate. Note that no similar dependencies could be observed for the $1\ \mu\text{m}$ sized beads due to the noise level and limited resolution of the experimental set-up.

The data of Fig. 5 show that for small bead-substrate distances, there are two local energy minima associated with each magnetic cylinder, located close to the left and right edge of each stripe. These minima wash out and get shallower for larger z_B values. For bead-substrate distances relevant for the present experiments ($z_B \geq 2.4\ \mu\text{m}$), there is only one energy minimum associated with each magnetic cylinder. This is the situation for which the present simulations are carried out. As the bead-substrate distance changes during the SPB motion, this has also been considered in the simulations (see below). The exact positions and depths of the energy minima depend on the distance between SPB and the substrate, z_B (Fig. 5), and their asymmetry increases with decreasing z_B (in Fig. 5, we show the results of calculations with a ten times smaller diameter than that used in the experiments, i.e., $0.4\ \mu\text{m}$ instead of $4\ \mu\text{m}$, to illustrate the dependencies for low elevation values z_B). The periodically changing external magnetic field modulates the $E(\vec{r})$ dependence by a stepwise lateral shift of the energy minima.^{8,12–14} In

FIG. 4. (a) Exemplary experimental time dependencies of the $4\ \mu\text{m}$ sized SPB radial position when moving at different angles ϕ with respect to the x -axis of the coordinate system. The dependencies correspond to the four xy -trajectories shown in Fig. 2 (Multimedia view) (the yellow, red, and green curves correspond to three trajectories of appropriate color that ends closest to the center of the cylinders in Fig. 2). (b) Average SPB velocity as a function of the external fields' driving frequency for $\phi \approx 0^\circ$ determined from the distance the SPBs covered within a time span of 2 s. The data point for each frequency refers to a single experimental trajectory, where the indicated error was derived from the uncertainty in the SPB trajectory.

FIG. 5. Calculated radial coordinate dependence of the $0.4\ \mu\text{m}$ sized SPB's magnetic potential energy landscape based on magnetic dipole approximation of the SPB's magnetic moment ($\phi = 0^\circ$). The results are shown for different heights z_B of the model bead center over the substrate surface. The external field in the calculation was $H_x = 1.2\ \text{kA/m}$ and $H_z = 1.6\ \text{kA/m}$, and the susceptibility of the SPB was set to 0.4. The z_B values are $2.4, 2.0, 1.8, 1.5, 1.2, 0.8, 0.5$, and $0.3\ \mu\text{m}$. The gray-shaded stripes indicate the r -positions of the magnetic stripes. Note that over the stripes, for $\Delta z > 0$, $E(r, z_B) < E(r, z_B + \Delta z)$ for each pair of curves.

Fig. 6, an explanation of the SPBs' directed motion is given. After allowing the SPB to approach the minimum of potential energy,³⁸ the external field is turned on (note again that the switching time is short in comparison to all other relevant timescales). Choosing the period of constant external field long enough, the SPB moves to the new position of the local minimum [curve 1 in Fig. 6(a) at $x \approx 32.5\ \mu\text{m}$] and rests there. Then, the direction of H_{ext} is changed and the SPB continues its motion towards the shifted potential energy minimum (curve 2).³⁹ Note that the SPB in this situation finds itself at a position where $E(\vec{r})$ spatially changes, and due to

FIG. 6. Calculated dependencies of the energy of a $4\ \mu\text{m}$ SPB in the dipole approximation on its r -coordinate for various orientations of the external magnetic field ($z_B = 2.4\ \mu\text{m}$): (a) for $\phi = 0^\circ$ and (b) for $\phi = 75^\circ$. The components of the external field in the calculation were $H_x = \pm 1.2\ \text{kA/m}$ and $H_z = \pm 1.6\ \text{kA/m}$. The curves 1–4 correspond to a clockwise change of the external field direction from $([+H_x, +H_z], [+H_x, -H_z], \dots)$. The gray-shaded stripes indicate the r -ranges of the magnetic stripes, and the dashed vertical line in (a) is placed at the minimum energy r -position of curve "1."

this, the gradient experiences a force in the $+x$ direction. Provided that the periods of constant external field are not too short ($v_{te} \cdot \frac{1}{4}T \sim w$, where T is the period of the external field variations and w is the width of the stripes), the SPB can reach the neighboring energy minimum (label “2”). Consecutive cyclic changes of the field direction transport the SPB to the second minimum of the dashed curve in Fig. 6(a), i.e., by approximately one spatial period of the underlying magnetic pattern—a particle transport mode referred to as phase-locked motion.⁴⁰ As seen in Figs. 1 and in 2 of the [supplementary material](#), the pattern of motion is qualitatively the same for different SPB sizes (1 and $4\ \mu\text{m}$ diameters) and in both the movies shown there. For the trajectory shown in Fig. 1 ($\phi = 0^\circ$), the z -coordinate of the SPB while it moves over the magnetic substrate changes within approximately $2.7\text{--}3.1\ \mu\text{m}$ and the maximal magnetostatic forces corresponding to these extremes are $2.5 \times 10^{-12}\text{N}$ and $1.8 \times 10^{-12}\text{N}$, respectively. Note that the behavior described so far is not characteristic of the specific pattern we used for creating the static energy landscape. It has been observed in many magnetophoretic systems with translational symmetry^{12–14} and is similar to the prevailing ratchet motion of microscopic systems.^{39–41} Figure 7 shows the position of SPB vs. time for three different initial positions. The pattern of motion is similar in all cases: cyclic changes of the field direction forward the beads to consecutive energy minima visible as plateau like regions within the overall SPB trajectory. After several cycles and depending on the field rotation frequency f , the SPB reaches the outer or the inner perimeter of the magnetic substrate where it becomes trapped at the edge minimum of the corresponding energy landscape and starts to oscillate (see Fig. 7). Within the central area, the SPBs move to the right. As magnetostatic forces cannot be the origin of bead motion in the center of the structure (the action range of these forces is of the order of $2\ \mu\text{m}$ from the edges of the curved stripes—see the trajectories in Fig. 7), they are moved forward by being pushed from incoming beads, possibly also involving hydrodynamic forces due to the velocity field around moving SPBs.^{42,43} The SPBs form chains under the action of the external field with a component that is parallel to the x -axis.⁴⁴ The chains move to the right and are continuously ripped apart by the magnetostatic forces at the edge of the inner cylinder. Figure 8 illustrates the most interesting property of the current investigation: the focusing or lensing of the SPB trajectories towards the central region of the nested cylinders [Fig. 2 (Multimedia view)]. The model describes the lensing effect for a broad range of initial bead positions excluding only trajectories with $\phi \approx 90^\circ$. This behavior can be understood from Fig. 6(b). As the angle ϕ of the SPB trajectory (starting at $x=0\ \mu\text{m}$) approaches 90° , the asymmetry along the x -coordinate in the energy landscape vanishes, becoming completely symmetric, and therefore, a preferred motion in the $+x$ -direction changes into statistically distributed forward-backward motions of the beads. Moreover, the energy landscapes approach degeneracy which results at $\phi = 90^\circ$ in a reduction of the 4 differing MFLs for the 4 external field directions into only two differing MFLs.

FIG. 7. Characteristic $r(t)$ dependencies calculated for $4\ \mu\text{m}$ SPBs placed initially at $z=2.4\ \mu\text{m}$ and for three different trajectory angles ϕ (see the corresponding curves in Fig. 8). In the simulation, the external magnetic field was turned on at $t=10\ \text{s}$, and the field sequence ($H_x = \pm 1.2\ \text{kA/m}$; $H_z = \pm 1.6\ \text{kA/m}$) was repeated with a frequency of $f=1\ \text{Hz}$. The arrows show the direction of the external magnetic field, within the xz -plane, at various stages of the simulation. The lower dashed curve illustrates the SPB oscillations obtained for $\phi = 120^\circ$ and t between $15\ \text{s}$ and $19\ \text{s}$.

FIG. 8. (a) Simulated in-plane trajectories of the $4\ \mu\text{m}$ sized SPBs initially placed at various positions (marked with small circles) and at the same height over the substrate ($z_B = 2.4\ \mu\text{m}$). The gray annuluses signify the positions of the radially symmetric magnetic pattern. Some of the trajectories (with initial $\phi = 0^\circ, 60^\circ, 75^\circ, 120^\circ$) are also shown in Figs. 1 and 7. The frequency of the external magnetic field was chosen to be 1 Hz. (b) Steady state-velocity, v_{te} , determined from the dependencies shown in panel (a) (dots) and Fig. 12(b) (squares), as a function of the initial angle ϕ of the SPB's trajectory. The velocities were determined using a radial distance covered by the SPB within 0.01 s after the external field direction was changed to $(-H_x, H_z)$. Note that the dependencies correspond to different external field amplitudes and frequencies of its variations. The lines are guides for the eye and show a cosine function scaled by constants A and B.

The results of the simulations are compared with the experiments [Fig. 2 (Multimedia view)] in Figs. 9 and 10. Figure 9 shows that the simulations, by not including thermal kinetics,¹¹ did not predict the movement of SPBs perpendicular to the radius of the cylinders seen in the experiment (compare with the discussion in the last paragraph of Sec. IV). This becomes particularly clear for the trajectories with $\phi \approx 80^\circ$: the simulated trajectory crosses multiple times the gap between the fourth and the fifth cylinder with average radial velocity approaching zero, while at the same time, the SPB traverses over several cylinders in the experiment and

FIG. 9. Direct comparison between the experimentally observed (dark yellow dots) and simulated in-plane trajectories of the SPBs with a diameter of $4\ \mu\text{m}$. The experimental trajectories of $4\ \mu\text{m}$ sized SPBs are shown in Fig. 2 (Multimedia view), too. In the simulations, the bead started in an xy -position close to the one seen in the experiment and with a height over the substrate $z_B = 2.4\ \mu\text{m}$.

FIG. 10. Comparison between the experimental (red dots) and corresponding simulated time dependencies of $4\ \mu\text{m}$ SPB radial coordinate r . The two pairs of curves are the same as those shown in Fig. 9. Note that the experimental starting positions of SPBs do not coincide exactly with those seen in the simulations as there the SPBs not always reach the equilibrium position before the field was switched on. The inset again shows the curves for $\phi \approx 302^\circ$, but this time the experimental dependence is shifted to the right by 1.33 s (red circles) and to the left by 1.67 s (gray circles) relative to the simulated one (dashed trajectory) to allow for a more direct comparison of the fine structure: in both curves, the SPBs occasionally move backwards, resulting in a reduced average velocity (compare Fig. 12).

slightly changes its direction of motion, ϕ_i , which is not reproducible in our calculation. For initial angles ϕ_i closer to 0° or 180° , the simulated and tracked trajectories diverge much less. The simulated and tracked $r(t)$ dependencies (Fig. 10) are qualitatively similar and are characterized by pairwise comparable average radial velocities (slope). For $\phi_i \approx 170^\circ$, the tracked dependence shows prominent changes of the instantaneous velocity every 0.25 s, i.e., whenever the field direction was changed. For $\phi_i \approx 302^\circ$, the experimentally observed decrease in the average velocity seems to corroborate the predictions from the simulations that occasionally the instantaneous velocity of the SPBs changes the direction when the external field is too weak or changes too frequently (inset of Fig. 10). The two experimental minima seen in the inset, although much smaller than the uncertainty of the measurement, are spaced exactly 3 s apart, implying their relation to the changes in the external field direction.

As usual for magnetophoretic systems based on a combination of periodically arranged local field sources and external field pulses of frequency f , the SPBs will display directed motion as long as their steady-state velocity v_{te} is high enough to surpass the distance between two successive locations of potential energy minima within the time period of constant field.^{13,40} Therefore, there is a critical frequency, f_c , of the external field pulses where the SPB can no longer follow the changing potential energy landscape (Fig. 11). There is a huge difference between the measured experimentally $v_{te}(f)$ dependence (Fig. 4) and the results of the simulation (Fig. 11), where the critical frequency seems to be underestimated by the model for the $4\ \mu\text{m}$ sized beads. This deviation is due to several reasons that will be discussed in the following, which, however, do not invalidate our theoretical model. The most significant impact is related to the simplifications which were introduced to reduce the computational effort. This becomes particularly severe for the point dipole approximation for the SPB magnetic moment (see Sec. III and [supplementary material](#)), which breaks down for the $4\ \mu\text{m}$ SPBs. In this case, the magnetic content is distributed within a sphere shell of a diameter close to that of the SPB.¹⁹ Since the bead size is comparable to the length scale of the magnetic structure of the substrate, simultaneous interaction with neighboring stray field sources is expected. Note that magnetostatic force included in the model also strongly depends on the actual distance between the magnetic volume and the magnetic substrate (Fig. 5), and so, the dipole approximation we use throughout the calculations strongly underestimates the interaction of the patterned substrate with the superparamagnetic content that is close to the Co layers.

As the external force on the SPBs depends on both the field and the field gradient, both quantities can be used to modify v_{te} and therefore the critical frequency. In the current

FIG. 11. Simulated $r(t)$ dependencies (for initial $\phi = 0^\circ$ and H values like in Fig. 7) for $4 \mu\text{m}$ sized SPBs for different frequencies of the external field variations ($f = 1, 1.5, 2, 2.5, 3, 8, 12$ Hz). The inset shows the f -dependence of the average velocity of the bead (determined from a distance covered in the time range between $1/4f$ and $5/4f$ s or between $10/f$ and $15/f$ s for $f \geq 8$ Hz). The change in a slope of the 2.5 Hz curve at $t \approx 11.5$ s is due to the SPB reaching the edge of the outer cylinder.

micromagnetic structure/field pulse arrangement, the magnetic field gradient depends on the angle ϕ of the SPB trajectory, and therefore, v_{te} will also depend on ϕ . Figure 8(b) shows v_{te} as a function of ϕ calculated for external field pulses of $H_x = \pm 1.2$ kA/m and $H_z = \pm 1.6$ kA/m and a frequency of 1 Hz and for twice that values. In the both cases, the decrease in v_{te} with increasing ϕ is obvious and can be well approximated with a cosine function, reflecting the ϕ dependence of the external field component parallel to the trajectory. A directed transport is only then possible when the SPBs will be able to surpass a distance larger than approximately half of the distance between successive potential energy minima, and there will be only a defined range $\Delta\phi$ of ϕ , where directed transport of SPBs is observed. This effect can be seen when comparing dependencies for $\phi = 0^\circ$ and $\phi = 75^\circ$ shown in Figs. 7 and 12(a): it is obvious that for higher f , the SPBs with higher ϕ are transported with lower effective velocities

FIG. 12. Trajectories of $4 \mu\text{m}$ sized SPBs calculated for four different initial angles ϕ ($0^\circ, 45^\circ, 60^\circ$, and 75°) for a fixed frequency of 2 Hz and two different values of the external magnetic field: (a) (H_x, H_z) and (b) $2 \times (H_x, H_z)$. The gray rectangle indicates one of the time ranges in which the SPB travels thrice over the same region (compared to the inset of Fig. 10).

(the SPB moving parallel to the external field can still move in the phase-locked manner). Therefore, by changing f , a weaker or stronger focusing of trajectories can be achieved. Similarly, as seen in Fig. 12, the angle dependence of the SPB velocity can be influenced by the external field amplitude.

A considerable part of microfluidic magnetophoretic systems operate under fluid flow. Within a more generic framework of microfluidic applications, the integration of a magnetophoretic transport system needs to be combined with the present hydrodynamic flow conditions. For the presented concept of magnetophoretic lensing, applying a flow parallel to the x -axis, i.e., parallel to the plane of the external driving field, would lead to a superimposed velocity component with respect to the magnetic field induced transport. This becomes particularly severe within regions where the potential energy landscape is degenerated along the x -axis, so that the intended motion perpendicular to the edges of the cylinders is disturbed [see e.g., the upper trajectory in Fig. 2 (Multimedia view)]. An appropriate solution to this problem might be to slightly modify the geometry of the cylinder pattern, e.g., by removing two opposing circle segments of several arcs with the width close to $\phi = 90^\circ$ and 270° to interrupt or prevent the presence of energetically degenerated potential wells. As a side-effect, the SPBs will be attracted by the edges of the cylinders as indicated in Fig. 5. Furthermore, since the transport takes place in close proximity to the substrate surface, the flow velocity is significantly reduced in comparison to the maximum flow velocity present in the central region of the channel, which could be of further advantage for the integration into microfluidic systems.

V. CONCLUSIONS

We have shown that the Co/Au multilayer with perpendicular magnetic anisotropy, which is topographically patterned into a set of concentric hollow cylinders, can be used for magnetophoretic lensing of SPBs. The modeling of the SPB trajectories corroborates that this feature is due to the curvature of the edges of the magnetic cylinders and demonstrated their suitability for spatially directed magnetophoresis. An alternative structure consisting of two adjacent 180° annulus sectors with opposite magnetizations could be perspective used to concentrate SPBs in the center of the pattern. In summary, the outlined experiments give rise to the use of a more general approach for magnetophoretically driven bioassays, in which the formation of SPB hybrids in spatially localized traps can be applied for biomolecular interaction analysis.

SUPPLEMENTARY MATERIAL

See [supplementary material](#) for the exemplary movies showing the movement of SPBs and some additional data.

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APPENDIX A: FORMULAS FOR CALCULATING RADIAL FIELD COMPONENTS

We cite the equation that we used to calculate the radial component of the magnetic field produced at point (r, z) by one surface of a cylinder (charged monopolar annulus symmetric about the z -axis, its surface is lying at z -coordinate h and is parallel to the xy -plane)—Eq. (4) from Ref. 24. The inner and outer radii of a cylinder are r_{in} and r_{out} , respectively. Its magnetization is M_s . The equation reads

$$H_r(r, z) = \frac{M_s}{2\pi} \frac{\sqrt{(r_{out} + r)^2 + (z - h)^2}}{r} \times \left(\left(1 - \frac{k_1^2}{2}\right) K^*[k_1] - E^*[k_1] \right) - \frac{M_s}{2\pi} \frac{\sqrt{(r_{in} + r)^2 + (z - h)^2}}{r} \times \left(\left(1 - \frac{k_2^2}{2}\right) K^*[k_2] - E^*[k_2] \right), \quad (A1)$$

where

$$K^*[k] = \int_0^{\phi=\frac{\pi}{2}} \frac{d\theta}{\sqrt{1 - k^2 \sin^2(\theta)}}, \quad (A2)$$

$$E^*[k] = \int_0^{\phi=\frac{\pi}{2}} \sqrt{1 - k^2 \sin^2(\theta)} d\theta \quad (A3)$$

are elliptic integrals and

$$k_{1(2)} = \frac{2\sqrt{r_{out(in)}r}}{\sqrt{(r_{out(in)} + r)^2 + (z - h)^2}}. \quad (A4)$$

APPENDIX B: FORMULAS FOR CALCULATING AXIAL FIELD COMPONENTS

Here, we cite the equation that we used to calculate the z -component of the magnetic field produced by a solid cylinder of radius r_{out} , with the z -axis of symmetry, and height h [Eq. (16) from Ref. 25 (also see Ref. 26)] at point (r, z) . The cylinder is magnetized axially with saturation magnetization M_s . The equation reads

$$H_z(r, z) = -\frac{M_s}{4\pi} \int_0^{2\pi} \int_0^{r_{out}} \left(\frac{R(h/2 - z)}{(R^2 + (h/2 - z)^2 + r^2 - 2Rr \cos \Phi)^{3/2}} + \frac{R(h/2 + z)}{(R^2 + (h/2 + z)^2 + r^2 - 2Rr \cos \Phi)^{3/2}} \right) dR d\Phi. \quad (B1)$$

To calculate the field of a hollow cylinder, one combines two equations of Eq. (B1) type.

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