Patterned time-orbiting potentials for the confinement and assembly of magnetic dipoles

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We present an all-magnetic scheme for the assembly and study of magnetic dipoles within designed confinement profiles that are activated on micro-patterned permalloy films through a precessing magnetic field. Independent control over the confinement and dipolar interactions is achieved by tuning the strength and orientation of the revolving field. The technique is demonstrated with superparamagnetic microspheres field-driven to assemble into closely packed lattice sheets, quasi-1D and other planar structures expandable into dipolar arrays that mirror the patterned surface motifs.

The confinement and manipulation of charge carriers within intricate low-dimensional structures through designed electric potentials have led to revolutionary advances in semiconductor devices. In contrast, the magnetic analog of creating designable energy landscapes for confining magnetic dipoles with concomitant tunability of the relevant forces solely through magnetic fields has however remained elusive. Such field-based approaches could offer powerful means to probe mesoscopic magnetic systems characterized by length scales ranging from tens of nanometers to micrometers, forces extending from femto- to nano-newtons and times scales above a microsecond.

Schemes to confine interacting magnetic dipoles of micrometer to centimeter scale have been proposed as early as in 1878. However, existing approaches confront challenges such as hard-wall confinements that prohibit regulation of the size of the dipole cluster, restriction of magnetic dipolar interaction to either repulsive or attractive, and lack of control on introducing and removing a specific number of dipoles (smaller than millimeter size) within the interacting landscape. Magnetic traps have also been developed for the confinement of dipolar atoms; however, the current-carrying coils utilized offer limited configurations on the spatial design and size of the trap.

Here we present a straightforward mechanism that overcomes these challenges by activating a time-orbiting potential on a micro-patterned permalloy thin film with a precessing magnetic field. Independent tunability of the soft-confinement force and dipolar interactions (reversibly between attraction and repulsion) as well as in-situ control on the number of confined dipoles are obtained solely through tuning the magnetic field. Moreover, the lithographically defined confinement scheme allows complex designer landscapes of desired length scales to be readily fabricated. Demonstrated by the remote, field-driven assembly of dipoles consisting of fluid-borne microspheres, this all-magnetic scheme serves as a prototype to stabilize various low-dimensional spatial constructs realizable over wide length scales. These features enable fundamental studies such as artificial atoms, nucleation, jamming and frustration as well as applications in biomedicine, material assembly, photonics, magnetic logic, chip based devices and atom traps.

Results

The architectures of the confining potential for magnetic dipoles are defined by permalloy (Ni₀.₈Fe₀.₂) thin-film patterns imprinted on a silicon substrate through photo or electronbeam lithography. Figure 1(a) schematically illustrates a random distribution of superparamagnetic microspheres (8 µm in diameter) on a permalloy disk pattern in solution that have, in the absence of magnetic fields, been drawn to the surface by gravity. An externally applied field $H_{ext} = (H_x, H_y, H_z)$ transforms the microspheres into dipoles while the permalloy disk becomes uniformly magnetized along the $(H_x, H_y, H_z)$ direction. The dipoles interact with each other as well as fields emanating from the disk, arranging themselves into configurations that minimize the total energy $U$, that has primary

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experiences an asymmetric energy landscape, 

Therefore, precessing

Figure 1 | Formation of a time-orbiting potential on a permalloy disk. (a) Illustration of magnetic microspheres randomly distributed atop a permalloy disk within fluid environment on a silicon substrate in the absence of external magnetic fields. (b) In the presence of \( H_{\text{ext}} = (60, 0, 60) \times 10^4 \) A/m, shown from top to bottom are the calculated energy landscape \( U_{\text{disk}} \), simulation, and experimental snapshot of the system in equilibrium. Contour interval on the energy landscape is 2000 \( k_B T \), where \( k_B \) is Boltzmann’s constant and \( T = 300 \) K the temperature. Projections of the microspheres on the surface are drawn in circles in the simulation to show levitation of microsphere chains along \( H_{\text{ext}} \). Scale bar in the snapshot is 100 \( \mu \)m. (c) Same as in (b) but with \( H_{\text{ext}} \) precessing about the \( z \)-axis at 20 Hz with in-plane component \( H_1 = (60,0) \times 10^4 \) A/m and angle of precession \( \theta = 45^\circ \), leading to a time-averaged energy landscape, \( U_{\text{disk}} \).

Figure 2 | Quantification of the two primary forces in the system: time-averaged confinement force \( F_{\text{conf}} \) and dipolar force \( F_{\text{dip}} \). In the presence of a precessing \( H_{\text{ext}} \) as in Fig. 1(c), the in-plane \( x \)-component of (a) \( F_{\text{conf}} \) and (b) \( F_{\text{dip}} \), (exerted by another microsphere located at \( x = 0 \)) are plotted as a function of \( x \) (in units of the disk radius \( R_{\text{disk}} \) and microsphere radius \( R \)). \( F_{\text{conf}} \) and \( F_{\text{dip}} \) are defined as the maximum in-plane confinement and dipolar forces respectively. (c) Contours of constant \( F_{\text{conf}}^* \) and \( F_{\text{dip}}^* \) are plotted in the parameter space spanned by \( H_1 \) and \( \theta \).

\[
F_{\text{conf}} = -\nabla U_{\text{conf}},
\]

that arise from the dipole-disk interaction and the dipolar force, \( F_{\text{dip}} = -\nabla U_{\text{dip}} \). In the presence of a precessing \( H_{\text{ext}} \) oriented at, for example, \( \theta = 45^\circ \) from the \( z \)-axis with an in-plane component \( H_1 = 60 \times 10^4 \) A/m, the confinement force experienced by a microsphere atop the disk is \(< 1 \) pN directed towards the center as shown in Fig. 2(a). An interesting property is the sole dependence of the confinement force on \( H_1 \), since \( F_{\text{conf}} \propto H_1 \). Figure 2(b) shows that under the same precessing field, the corresponding dipolar force between two microspheres on the surface is \(~0.2\) pN (repulsive) and weakens with their separation \( x \) as \( 1/x^4 \). Moreover, since \( F_{\text{conf}} \propto H_1^2 (2 \cos^2 \theta - 1) \), the dipolar force can be tuned from being repulsive \( (\theta < \theta_c ) \) to attractive \( (\theta > \theta_c ) \), crossing over at \( \theta = \theta_c = \cot^{-1} \left( \frac{1}{\sqrt{2}} \right) = 54.7^\circ \). Separate and independent control on the confinement and dipolar forces can thus be obtained, as illustrated in Fig. 2(c), by tuning \( H_1 \) and \( \theta \) along contours of constant \( H_1 \) or constant \( H_1^2 (2 \cos^2 \theta - 1) \). Derivations for the above, including the expressions:

\[
U_{\text{conf}}(r) = -\frac{\mu_0 H_1}{8 \pi} |r| V M_{\text{disk}} d\left( 2 - m - 4 \frac{R_{\text{disk}}^2}{\rho^2} \right) I_0(m, m) - 2K(m),
\]

and

\[
U_{\text{dip}}(r) = -\frac{\mu_0}{8 \pi} |r|^2 V^2 H_{\text{disk}}^2 (3 \cos^2 \theta - 1) \sum_{r \neq r} \frac{3 \cos^2 \theta - 1}{|r - r'|^3}
\]

are available as Supplementary Information.
The ability to tune the dipolar interaction (with $\theta$) while the soft-confinement force is fixed (with $H_1$) allows the confined dipoles to assemble into various structures. An important parameter characterizing the dipole structure is the average center-to-center microsphere separation, $\langle \Delta r \rangle$, determined by Delaunay triangulation which serves as a measure of the cluster size. As shown in the plot of Fig. 3, $\langle \Delta r \rangle$ is varied over a factor of two from $4R$ to $2R$ (in contact) as $\theta$ is increased from $35^\circ$ to $70^\circ$. In the repulsive regime of dipolar interaction ($\theta < \theta_c$), the dipoles form an expanded cluster within a center-attracting potential (insets A, B, C, F and G of Fig. 3), resembling the two-dimensional version of J. J. Thomson’s atomic “plum pudding model” and lending itself as an unstudied form of artificial bbling the two-dimensional version of J. J. Thomson’s atomic ''plum center-attracting potential (insets A, B, C, F and G of Fig. 3), resem-

Discussion

The time-orbiting bowl-shaped profile can be readily extended to other shaped confining potentials with surrounding barriers and trenches defined by permalloy patterns. Shown in Fig. 4(a–d) are examples of triangle, square, long rectangle and octagonal ring geometries that maintain the same independent control on the confining and dipolar forces through $\theta$ and $H_1$. These energy landscapes unveil...
previously unexplored opportunities to study field-driven assembly of magnetic dipoles beyond conformations with cylindrical symmetry or simple topology. For instance, the dipole cluster expands to reflect different trap motif and symmetries under repulsive dipolar interplay; the long rectangle serves as a quasi 1D magnetic channel; the discrete clusters decorating the octagonal ring is a result of the rare competition among inter-particle attractive force and the confinement force – two forces that are typically cooperative. The intricate balance between the interplaying forces and geometry of the soft-confining potential leads to the emergence of a complex and design-specific behavior of the dipole clusters.

The advantages of the all-magnetic approach presented here include easy, non-contact manipulation of the dipoles by weak external magnetic fields, and absence of intricate wiring patterns for addressing specific electrodes as well as screening and heating effects that are generally present in charge-based approaches. We expect the presented scheme to have useful applications, for instance, in biomedical devices11–13: control of local concentration effects that are generally present in charge-based approaches. We expect the presented scheme to have useful applications, for instance, in biomedical devices11–13: control of local concentration of magnetic dipoles beyond confinements with cylindrical symmetry and optimal mixing can be achieved by the expansion and collapse of the dipole cluster; attracting the dipoles from different directions to the center of the disk serves as a simultaneous, multi-directional force probe on a biological entity attached to the center; dipole clusters of tunable spacing can also be fixed as filters to sort out objects of different sizes within a flow channel. From a materials stand-

### Microsphere solution

Superparamagnetic microspheres (UCMCF-9560, Bangs Laboratories) with diameter of 8 μm are diluted in de-ionized water with 0.01 ~ 0.05% Tween-20 (Sigma-Aldrich) for use on spin-coated SiO2 surface or with 0.02 ~ 0.05% Triton X-100 (Sigma-Aldrich) for use on magnetron-sputtered SiO2 surface, preventing surface adhesion. The diluted microsphere solution is then placed on the permalloy-patterned substrate and contained in a polydimethylsiloxane ring capped by a cover glass to eliminate evaporation and fluid flow.

### Magnetic field generation

Computer program (LabVIEW, National Instruments) controls the current sent by the power sources (BOP 20–10 M, Kepco) into the electronics and solenoid, providing three-dimensional and optimal control of the external magnetic field Hext applied on the sample. To prevent net rotation and drifting of the microsphere cluster when a precessing field about the z-axis is present, the rotation of the in-plane field is programmed to cycle through the following sequence of angles at a speed of 20 revolutions per second: 180°, −360°, 180°, −360°, 360°, −180°, 360° and −180°. Note that the time period is 0.3 s per cycle.

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**Methods**

**Fabrication of permalloy thin-film patterns.** Permalloy thin-film patterns are imprinted on a silicon substrate by standard photolithography and magnetron-magnetron lithography: For the disk pattern, two layers of photo resist (LOR3B and Si1805, MicroChem) are applied to the Si substrate through spin-coating at 2500 rpm followed by baking at 180 °C and 110 °C on a hotplate for the respective layers; the disk patterns are then exposed through an optical microscope (BH-2, Olympus) and developed (in MF-319, Dow). For other patterns, two layers of e-beam resist (methylmethacrylate and polyethyl methacrylate, MicroChem) are applied to the Si substrate through spin coating at 4500 rpm followed by baking on a 180 °C hotplate; pattern areas are exposed at 125 μC/cm² using a scanning electron microscope (Helios Nanolab 600, FEI Company) and developed using 1:3 methyl isobutyl ketone isopropyl alcohol (MicroChem). A 40 nm layer of Ni80Fe20 (permalloy) is magnetron-sputtered onto the resist-patterned substrate by magnetron sputtering (ATC, Orion, AJA International) followed by lifing off unwanted areas with acetone and 60 °C bath of n-methyl-2-pyrrolidone. The resulting permalloy patterns are protected from the environment by another layer of SiO2 (100 ~ 200 nm) through spin coating (Silicafilm, Emulsitone) at 4000 rpm and baking at 180 °C for the disk patterns, or through magnetron sputtering for other patterns. The spin-coated SiO2 surface is treated with NaOH (0.1 M) for 10 min prior to experiment. The dimensions of various patterns are (in μm): circular disk (210 in diameter), triangle (260 in side length), square (150 × 150), rectangle (75 × 300) and octagonal ring (200 × 250 with a 50 × 100 hole).
