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Resonantly excited precession motion of three-dimensional vortex core in magnetic nanospheres

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We found resonantly excited precession motions of a three-dimensional vortex core in soft magnetic nanospheres and controllable precession frequency with the sphere diameter $2R$, as studied by micromagnetic numerical and analytical calculations. The precession angular frequency for an applied static field H_{DC} is given as $\omega_{MV} = \gamma_{eff} H_{DC}$ where $\gamma_{eff} = \gamma \langle m_T \rangle$ is the effective gyromagnetic ratio in collective vortex dynamics, with the gyromagnetic ratio γ and the average magnetization component $\langle m_T \rangle$ of the ground-state vortex in the core direction. Fitting to the micromagnetic simulation data for $\langle m_T \rangle$ yields a simple explicit form of $\langle m_T \rangle \approx (73.6 \pm 3.4)(l_{ex}/2R)^{2.20 \pm 0.14}$, where l_{ex} is the exchange length of a given material. This dynamic behavior might serve as a foundation for potential bio-applications of size-specific resonant excitation of magnetic vortex-state nanoparticles, for example, magnetic particle resonance imaging.

The Larmor precession is a universal dynamic phenomenon in nature that represents the precession of a magnetic moment about a magnetic field at a characteristic Larmor frequency, which is expressed as $\omega_L = \gamma H$, where γ is the gyromagnetic ratio and H , the static field strength. This type of precession plays very crucial roles in a rich variety of electron- or nuclei-spin-related dynamics such as electron-spin resonance, nuclear magnetic resonance, ferromagnetic resonance, and related magnetization dynamics^{1–6}. Such dynamic fundamentals have been widely utilized in a significant number of applications, including material analysis^{1,4}, bio-medical imaging^{7,8}, and information recording in magnetic media^{9,10}.

In this paper, we report the discovery of resonantly excited precession motions of a magnetic vortex core in soft magnetic nanoparticles of spherical shape¹¹, but with totally different underlying physics from those for vortex motions so far reported^{12–17}. We also were able to identify sphere-size-controllable precession angular frequency ω_{MV} and size-specific resonant excitations of nanoparticles bearing a magnetic vortex structure. We additionally determined, based on combined micromagnetic numerical and analytic calculations, that the size specificity of ω_{MV} originates from the variable effective gyromagnetic ratio with the sphere size that modifies the vortex structure inside spheres. Our results could provide a potential means of implementing size-specific resonant excitation of nanoparticles in bio-applications¹⁸.

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Figure 1. Ground-state magnetization configurations of Py nanospheres according to the diameter. (a) Finite-element sphere model for diameter $2R = 30$ nm. (b) Ground-state magnetization configurations of Py nanospheres for different $2R$ values as indicated: upper, viewed from positive z -direction and sliced across x - y plane; lower, viewed from positive x -direction and sliced across y - z plane. The color represents the x -component of the local magnetizations, $m_x = M_x/M_s$ (see the color bar). The arrows inside the sphere of $2R = 150$ nm represent the local curling magnetizations. (c) m_x profiles along y axis for different diameters. The inset shows the m_x profiles versus the normalized distance for each diameter.

Results

Ground states of nanospheres. Figure 1a shows a nanosphere model of spherical symmetry. As described in Methods, we performed micromagnetic numerical calculations on Permalloy (Py, $\text{Ni}_{80}\text{Fe}_{20}$) nanoparticles of different diameters, $2R = 10$ nm – 150 nm (see Methods). Figure 1b illustrates the ground states of the spheres obtained through relaxation from their saturated states in the $+x$ direction. For the $2R < 40$ nm cases, uniformly magnetized single-domain states were obtained, whereas for the $50 \text{ nm} \leq 2R \leq 150$ nm cases, single magnetic vortex states were well established. The vortex state of the $2R = 150$ nm sphere, for example, was visualized by streamlines circulating around the vortex core oriented in the $+x$ direction. We noted that the region of the in-plane circulating magnetizations varies markedly with $2R$, as indicated by the x -component of the local magnetization $m_x (=M_x/M_s)$ profiles in Fig. 1c. This dramatic variation is the result of a strong competition between the long-range dipolar and short-range exchange interactions in those nanospheres of such varying size.

Resonantly excited precession motion of a vortex core in spheres. Since the spherical symmetry of nanospheres does not lead to any magnetic shape anisotropy, when a sizable static field H_{DC} is applied in the $+z$ direction, the vortex cores for $40 \text{ nm} < 2R \leq 150$ nm start to reorient to the field direction, but with accompanying precession motions (see Supplementary Movie). This precession motion is different from the well-known gyration and even its higher-order modes of vortex cores in planar dots^{12–17}. Although very weak spin waves are emitted inside the nanospheres, the vortex's spin configurations are maintained as a whole structure, because the field strength is sufficiently small. In the relaxation process, the core orientation converges in the field direction ($+z$ -direction), reflecting the fact that the m_x averaged over the entire volume of the sphere, $\langle m_x \rangle$, undergoes decaying oscillation through its vortex-core precession (inset of Fig. 2a). The precession frequency was obtained by Fast Fourier Transformation (FFT) of the temporal $\langle m_x \rangle$ evolution for the different values of $2R$ and H_{DC} (see Fig. 2a,b, respectively). In the cases of uniformly saturated particles ($2R = 10, 20$, or 30 nm), the

Figure 2. Precession frequency of Py nanospheres as functions of (a) $2R$ and (b) H_{DC} applied in $+z$ direction (perpendicularly to initial vortex-core orientation). The inset in (a) shows the $\langle m_x \rangle$ oscillation versus time, for a sphere of $2R = 80$ nm. In (a), uniform single-domain (SD) and vortex states are distinguished at about $2R = 37$ nm by the gray color. The symbols indicate the micromagnetic numerical calculations, with corresponding lines drawn by eye. In (b), the lines are the results of linear fits for the individual diameters, as indicated.

frequency was independent of $2R$, as determined by the Larmor frequency $f_L = (\gamma/2\pi)H_{DC}$ ¹⁹. By contrast, for the vortex-state spheres ($40 \text{ nm} \leq 2R \leq 120 \text{ nm}$), the precession frequency of a vortex core showed a strong variation with $2R$, as can be expressed by $f_{MV} = (\gamma_{eff}/2\pi)H_{DC}$, where $\gamma_{eff} (< \gamma)$ is the effective gyro-magnetic ratio, which is variable with the sphere diameter.

In order to quantitatively elucidate the γ_{eff} -versus- $2R$ relation, we plotted the value of f/H_{DC} as a function of $2R$, as compared with the average magnetization component over the sphere volume in the vortex-core orientation, $\langle m_T \rangle$, both of which were obtained from the micromagnetic simulations. As shown in Fig. 3, when $\gamma/2\pi = 2.8$ (MHz/Oe) on the left axis is scaled to $\langle m_T \rangle = 1$ on the right axis, both numerical values are in excellent agreement over the entire range of diameters studied, resulting in an explicit form of $\gamma_{eff}/\gamma = \langle m_T \rangle$ (for single-domain states, $\gamma_{eff} = \gamma$, because of $\langle m_T \rangle = 1$). Therefore, the precession frequency of a vortex core in nanospheres can be expressed as $f_{MV} = (\gamma/2\pi)\langle m_T \rangle H_{DC}$. This precession frequency cannot be explained by the gyration mode (or even by higher-order modes) of vortex cores in thin or thick film dots, and neither, consequently, by Thiele's equation^{12–17}.

Analytical derivation of vortex-core precession in nanospheres. In order to gain physical insight into the $f_{MV} = (\gamma/2\pi)\langle m_T \rangle H_{DC}$ relation obtained from the micromagnetic simulations, we analytically derived vortex-core precession dynamics in nanospheres. In our modeling, a weak static field was applied in the $+z$ direction, which field sustained the rigid vortex structure in a certain potential, and thus allowed the initial ground-state vortex core to align in the $+z$ direction through the precession around the field direction along with certain damping. We used the local spherical reference frame on infinitesimal segments of the surface, where the unit vector of local magnetizations is expressed as $\mathbf{m} = (m_r, m_\theta, m_\phi)$, r is the radial distance, θ is the polar angle, and ϕ is the azimuthal angle, as shown in Fig. 4a. Time-variable vortex-core orientation can be defined as a unit vector $\mathbf{\Gamma} \equiv (\sin\theta_0 \cos\phi_0, \sin\theta_0 \sin\phi_0, \cos\theta_0)$, as illustrated in Fig. 4b. Following the rigid vortex Ansatz, which agreed with the micromagnetic simulation results, local magnetizations inside a given sphere could be expressed as $m_r = f(r, \mathbf{\Gamma} \cdot \hat{\mathbf{r}})$ and $\Phi = -\arctan_s(\mathbf{\Gamma} \cdot \hat{\mathbf{\theta}})/(\mathbf{\Gamma} \cdot \hat{\mathbf{\phi}})$, where Φ is the azimuthal angle of the magnetization in the local spherical reference frame (inset of Fig. 4a). Here we assume some general shapes of m_r that are restricted by the condition $f(r, 1) = -f(r, -1) = 1$ for all r values. Since m_r

Figure 3. Precession frequency normalized by H_{DC} (circles) and $\langle m_r \rangle$ (crosses) obtained from micromagnetic numerical calculations. The value of $\gamma/2\pi = 2.8$ (MHz/Oe) on the left axis is scaled to $\langle m_r \rangle = 1$ on the right axis. The different colors of the circle symbols indicate the numerical data for different sphere diameters, as indicated by the colors shown in Fig. 2a. The solid curve is the result of a numerical calculation of the analytical form of $\langle m_r \rangle \approx (73.6 \pm 3.4)(l_{ex}/2R)^{2.20 \pm 0.14}$.

Figure 4. Model for analytical derivations. (a) Definition of spherical coordinates and local spherical reference frame (colored surface) for local magnetization $\mathbf{m}$. (b) Schematic of model sphere wherein single rigid vortex core is pointed in direction of θ_0 and φ_0 , as defined by the polar and azimuthal angle coordinates.

and Φ are canonically conjugated variables, the time evolution of the local magnetizations can be determined from the Landau-Lifshitz-Gilbert (LLG) equations^{20,21} as

$$\dot{m}_r = -\frac{\gamma}{M_s} \frac{\delta E}{\delta \Phi} - \alpha(1 - m_r^2) \dot{\Phi}, \quad (1a)$$

$$\dot{\Phi} = \frac{\gamma}{M_s} \frac{\delta E}{\delta m_r} + \frac{\alpha}{1 - m_r^2} \dot{m}_r. \quad (1b)$$

By inserting the m_r distribution function of the vortex's spin configuration into Eqs. (1a) and (1b), we finally obtained the governing equation for vortex-core precession motion,

$$\dot{\mathbf{r}} + \frac{\gamma}{M_s V} \mathbf{r} \times \frac{\partial E}{\partial \mathbf{r}} + \frac{\alpha}{V} \mathbf{r} \times \frac{\partial F}{\partial \mathbf{r}} = 0 \quad (2)$$

where E is the total magnetic energy, F is a dissipative functional $\left[F = \frac{V}{2} \int d\mathbf{r} \left[\sin^2 \Theta \left(\frac{\partial \Phi}{\partial \mathbf{r}} \cdot \dot{\mathbf{r}} \right)^2 + \left(\frac{\partial \Theta}{\partial \mathbf{r}} \cdot \dot{\mathbf{r}} \right)^2 \right] \right]$, and $V = \frac{4}{3} \pi R^3$ is the sphere volume. The first, second and third terms in Eq. (2) correspond to the gyrotropic, potential energy and damping terms, respectively. The total energy E under a weak magnetic field applied along the z -axis, $\mathbf{H} = H_{DC} \hat{\mathbf{z}}$, can be expressed simply as $E_H = -\hat{\mathbf{z}} \cdot \mathbf{r}(t) H_{DC} V M_s \langle m_r \rangle$, where $\langle m_r \rangle$ is rewritten as $\langle m_r \rangle = \frac{1}{M_s V} \int d\mathbf{r} (\mathbf{M} \cdot \mathbf{r})$. Eq. (2) expresses the precession motion of vortex cores in collective spin dynamics; it differs from Thiele's equation to describe the gyration of vortex cores in planar dot systems.

By inserting E_H into Eq. (2) and assuming negligible damping, the precession frequency of a rigid vortex core can be given as $\partial \phi_0 / \partial t = 2\pi f_{MV}$ with $f_{MV} = (\gamma/2\pi) \langle m_r \rangle H_{DC}$. Consequently, we obtained the effective gyromagnetic ratio of the motion of a vortex in a given nanosphere as $\gamma_{\text{eff}} = \gamma \langle m_r \rangle$. This analytic form provides a clear physical insight into $2R$ -dependent f_{MV} , because $\langle m_r \rangle$, as indicated in the micromagnetic simulation results, varies with $2R$. Here we note that the eigenfrequency of a single vortex in cylindrical dots is known to vary with the aspect ratio of thickness L to $R^{12,15-17}$. However, the underlying physics of the size-dependent change in the precession frequency of the vortex core in nanospheres is totally different from that of the vortex gyration in planar disks, though both apparently show core-oscillation phenomena.

Dependence of $\langle m_r \rangle$ on sphere's diameter and constituent material parameters. Next, it is necessary to quantify how $\langle m_r \rangle$ varies with $2R$. We estimated, from further micromagnetic numerical calculations, the quantitative relation between $\langle m_r \rangle$ and $2R$ within the $2R = 50$ – 200 nm range for the different material parameters of both M_s and A_{ex} . Figure 5 reveals that $\langle m_r \rangle$ is given as $\langle m_r \rangle \approx \eta [A_{ex} / (2\pi M_s^2)]^{1.09 \pm 0.05} (2R)^{-2.21 \pm 0.13}$ with $\eta = 73.6 \pm 3.4$. According to the relation $l_{ex} = \sqrt{A_{ex} / (2\pi M_s^2)}$, $\langle m_r \rangle$ can be simplified as $\langle m_r \rangle \approx (73.6 \pm 3.4) (l_{ex} / 2R)^{2.20 \pm 0.14}$. This explicit form provides a simple and reliable estimation of $\langle m_r \rangle$ for a given value of $2R$ and a given material of l_{ex} , though there is yet no concrete model matching the form. We also note that, based on the single-domain states of $\langle m_r \rangle = 1$, the critical size for transition from a single domain to a vortex state^{1,22} can be simply estimated as $2R_c = 7.06 l_{ex}$. For example, the critical diameter, $2R_c = 37.3$ nm for Py, was in good agreement with that obtained from the simulation results shown in Fig. 1. As quantitatively interpreted, the strong variation of $\langle m_r \rangle$ versus $2R$ for a given material is related to the competition between the short-range, strong exchange interaction and long-range, but relatively weak dipolar interaction in nanospheres of given dimensions.

Size-specific resonant excitations. As an application of the aforementioned fundamental dynamics, we could activate magnetic nanoparticles of a specific size by tuning the frequency of an applied AC field to the f_{MV} of a sphere of a given diameter and material. In this modeling, an external AC field and a static field were given by $\mathbf{H}_{AC} = H_{AC} \sin(2\pi f_{AC} t) \hat{\mathbf{y}}$ and $\mathbf{H}_{DC} = H_{DC} \hat{\mathbf{z}}$, respectively, with sufficiently small values of $H_{AC} = 10$ Oe and $H_{DC} = 100$ Oe to avoid deformation of the initial vortex structures in the Py spheres. Figure 6a shows the oscillation of the core orientation θ_0 from the $+z$ direction during the precession process for $2R = 60$ nm ($f_{MV} = 95$ MHz), as excited by $f_{AC} = 91, 95$ and 99 MHz. The oscillation of θ_0 was hardly observable for the cases where f_{AC} was far from f_{MV} , whereas it was very large for the case of $f_{AC} = f_{MV}$, that is, at resonance. The resonantly excited precession leads even to vortex-core reversals between $\theta_0 = +\pi$ and 0 , as such reversals in planar disks occur periodically by linearly oscillating fields or currents applied on the disks' plane under the resonance condition^{3,23}. The oscillation of θ_0 represents a transfer of the external magnetic field to a magnetic sphere via the absorption of the Zeeman energy and subsequent emission to another form. The maximum energy absorption can be defined by the first maximum energy increment, ΔE_1 , as noted in Fig. 6a. Figure 6b plots ΔE_1 versus f_{AC} for different sphere diameters²⁴. For each diameter, the maximum peak height ΔE_{max} in the ΔE_1 -versus- f_{AC} curves was obtained under the corresponding resonance condition. All of the curves were well separated from each other, indicating reliable size-specific excitation of the magnetic particles. For example, the difference in f_{MV} between the 50 and 60 nm particles was about 50 MHz, which is sufficiently large compared with the full width at half maximums of both particles, 6.6 and 9.9 MHz, respectively.

In Fig. 6c are shown the ΔE_{max} -versus- $2R$ curves for comparison between the simulation data (solid circles) and the analytical form (lines) of the Zeeman energy, $\Delta E_{\text{max}} = 2H_{DC} V M_s \langle m_r \rangle$, where $\langle m_r \rangle = 1$

Figure 5. Calculation of $\langle m_r \rangle$ for indicated values of $2R$, $A_{ex}/A_{ex,Py}$ and $M_s/M_{s,Py}$. For each graph, one parameter is fixed: (a) $A_{ex}/A_{ex,Py} = 1$, (b) $M_s/M_{s,Py} = 1$, (c) and (d) $2R = 100$ nm. In both (a) and (b), $2R$ is in the 50–200 nm range. All of the symbols were obtained from the micromagnetic simulation results. The lines indicate linear fits.

for single-domain states or $\langle m_r \rangle \approx (73.6 \pm 3.4)(l_{ex}/2R)^{2.20 \pm 0.14}$ for vortex states. The simulation and analytical calculation agreed very well, as can be seen. The analytical calculation clearly shows that the magnetic energy absorption varies with $(2R)^3$ and $(2R)^{0.8}$ for the single-domain and vortex states, respectively. These results suggest that the magnetic energy absorption can be maximized by tuning f_{AC} to the resonance frequency of a given-diameter particle. This effect is made possible through size-specific resonance, size-selective activation and corresponding detection of the magnetic nanoparticles of a vortex state.

Discussion

We discovered, by micromagnetic numerical calculations, not only the resonantly excited precession motion of a vortex core in nanospheres and its size-dependent precession frequency, but also its physical origin, based on the size effect on the effective gyromagnetic ratio in collective spin dynamics analytically derived. This finding paves the way for size-selective activation and/or possible detection of magnetic nanoparticles by application of extremely low-strength AC fields tuned to the resonant frequency of a given diameter and material. These results, notably, would be applicable to magnetic particle resonance imaging (MPRI) and bio-applications.

Methods

In our micromagnetic numerical calculations, the FEMME code (version 5.0.8)²⁵ was used to numerically calculate the motions of the magnetizations of individual nodes (mesh size: ≤ 4 nm) interacting with each other via exchange and dipolar interactions at the zero temperature, as based on the LLG equation^{20,21}. The surfaces of the model spheres were discretized into triangles of roughly equal area

Figure 6. Total magnetic energy variation. (a) Total magnetic energy and polar angle of core orientation θ_0 during excitations of vortex core in sphere of $2R = 60$ nm by oscillating fields of $H_{AC} = 10$ Oe with different field frequencies ($f_{AC} = 91, 95$ and 99 MHz) under static field of $H_{DC} = 100$ Oe applied on $+z$ -axis. (b) Plot of ΔE_1 versus f_{AC} in $f_{AC} = 25$ – 310 MHz range. (c) Maximum absorption energy ΔE_{max} versus $2R$, calculated from micromagnetic simulations (solid circles) and analytical form (lines) of ΔE_{max} described in text.

using Hierarchical Triangular Mesh (HTM), as shown in Fig. 1a, in order to prevent irregularity-incurred numerical errors²⁶. The chosen material parameters corresponding to Py were as follows: saturation magnetization $M_s = 860$ emu/cm³, exchange stiffness $A_{ex} = 1.3 \times 10^{-6}$ erg/cm, damping constant $\alpha = 0.01$, $\gamma/2\pi = 2.8$ MHz/Oe, and zero magnetocrystalline anisotropy for the soft ferromagnetic Py material.

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Author Contributions

S.-K.K. and J.L. conceived the main idea and planned the micromagnetic simulation. M.-W.Y., J.L., H.-Y. L. and J.-H.L. performed the micromagnetic simulations and M.-W.Y., J.L. analyzed the data. Y.G., V.P.K. and D.D.S. derived analytical expressions for the precession motion of vortex core. S.-K.K. wrote the manuscript and all the coauthors commented on the manuscript.

Additional Information

Supplementary information accompanies this paper at <http://www.nature.com/srep>

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Corrigendum: Resonantly excited precession motion of three-dimensional vortex core in magnetic nanospheres

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The original version of this Article contained an error in the title of the paper, where the word “excited” was incorrectly given as “exited”. This has now been corrected in both the PDF and HTML versions of the Article.