Nanoparticle-modified magnetic vortex dynamics

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Abstract - With a view towards ferromagnetic-resonance-mediated detection of magnetic nanoparticles, we use micromagnetic simulations to study how the stray magnetic field produced by a uniformly magnetized spherical magnetic nanoparticle can modify the vortex gyrotropic resonance. We also show how these modifications depend on the particle's position and size. For small particle sizes, a core confinement effect induced by interaction between the particle's highly localized out-of-plane stray field and the vortex core magnetization can induce large frequency shifts. However, the generation of large shifts via this mechanism relies on the core's trajectory being localized directly below the particle. For larger particles (which generate stronger but less localized stray fields), changes in the gyrotropic frequency result from a combination of wide-scale out-of-plane spin canting and in-plane-field-induced shifting of the vortex core within its anharmonic confining potential. Compared to confinement-driven frequency shifting, the generation of large frequency shifts ($\gtrsim 10$ MHz) via these latter mechanisms is less dependent on particle positioning and the core orbit radius. The use of large particles thus represents a more reliable sensing modality if the particles cannot be guaranteed to align centrally over the disk.

Index terms - Nanomagnetism, vortex dynamics, micromagnetic models, magnetic sensors, biomagnetic particles.

I. INTRODUCTION

The magnetic vortex configuration arises naturally in (sub)micron sized ferromagnetic disks and consists of an in-plane curling magnetization that turns out-of-plane at the vortex's nano-scale core [Fig. 1(a)] [1], [2], [3], [4], [5]. Vortices exhibit a gyrotropic resonance which involves an orbit-like motion of the vortex core within its geometrically induced confining potential [6], [7]. Together with characteristics of the vortex spin structure, the core confining potential determines the frequency of core gyration with more highly confined (stiffer) cores tending to have a higher gyrotropic frequency, f_G [6]. Both the vortex spin structure and vortex core confinement can be modified by an external field, H [8], [9], [10], [11], [12], thus enabling field-tuning of f_G (f_G can also be modified by geometrical and magnetic inhomogeneities [13], [14], [15], [16]). The field-dependence of f_G has led to the use of vortices for tuneable electronic oscillators [17], [18], data storage [19] and frequency-based [20] magnetic field detection [21], [11].

One potential application of magnetic field detection is in magnetic biosensing for e.g., point-of-care medical diagnostics [22], [23]. In this biosensing approach, functionalized magnetic nanoparticles (MNPs) are used as labels for biological analytes. Exploiting the fact that most biological samples have a negligible magnetic background [24] the presence of these analytes can then be inferred by detecting the stray magnetic field created by the MNP tag(s). This detection is often

Fig. 1. (a) A simulated magnetic vortex magnetization configuration in a Permalloy-like disk with width ($2R$) 384 nm and thickness (L) 30 nm (zero magnetic field). The arrows show the direction of the in-plane curling magnetization (also encoded via color). The central white spot corresponds to the vortex core. (b) The simulation geometry used in this work: a ferromagnetic disk supporting a vortex together with an out-of-plane magnetized MNP (radius = R_P) which is modeled as a dipole magnetic field source located at a height, $d + R_P$, above the top surface of the disk.

achieved using Hall Effect sensors or conventional tunneling or giant magnetoresistive field sensors [23] (in the latter, MNP fields modify the quasi-static magnetization in the device's sensing layer, leading to a change in the device resistance [25]). MNP fields can however also modify vortex *dynamics* (and statics) [9], [11] and these dynamics can be probed electrically [17], [18], [26], opening pathways for vortex-based, electronic biosensors [21], [11]. Magnetic biosensing techniques that are based on (electrically) detecting changes in the frequency of resonant magnetization dynamics [20], [27], [28], [29] contrast the more conventional approaches outlined above in that they can operate naturally in the frequency domain (typically $\sim 0.1 - 10$ GHz). Other potential advantages of a frequency based approach include fast sensor response times (e.g., [30], [31]), highly promising size scalability [20] and a wide operational external field range [32], the latter enabling the generation of large particle moments [28].

In this paper we use micromagnetic simulations to study how the stray magnetic field produced by a uniformly magnetized, spherical MNP can modify the vortex gyrotropic resonance frequency. Specifically, we show how the observation of significant MNP-induced frequency shifts (~ 10 MHz or more) as well as the mechanisms underlying these shifts depends on particle size, particle position and core orbit radius.

II. MICROMAGNETIC SIMULATION METHOD

Simulations were run using MuMax3 [33] with the system geometry shown in Fig. 1(b). The disk (diameter $2R = 384$ nm and thickness $L = 30$ nm) is Permalloy-like: saturation magnetization $M_S = 800$ kA m $^{-1}$; exchange constant $A_{ex} = 13$ pJ m $^{-1}$; nil intrinsic magnetic anisotropy; damping constant $\alpha = 0.008$; gyromagnetic ratio $\gamma = 1.7595 \times 10^{11}$ rad(T.s) $^{-1}$ (within $\sim 5\%$ of the experimentally determined value [34]). A rectangular discretization with a cell size of $3 \times 3 \times 3.75$

nm^3 was employed. We will consider the case of a vortex with an anticlockwise chirality and a polarity aligned with the particle's magnetization ($p = +1$). Note the polarity can be set experimentally with an out-of-plane (OOP) magnetic field (e.g., [8]). The polarity also defines the sign of the change in f_G due to OOP fields [8].

The MNPs, MNP clusters or MNP-embedded polymer beads used in biosensing are typically superparamagnetic [23]. As such, an external field is required to obtain a finite time-averaged MNP magnetic moment which will be maximized in large fields. The OOP field geometry is preferable for vortex-based sensing in terms of maximizing the MNP moment without destroying the vortex state: the in-plane annihilation field is typically 10's of mT [1] whereas the out-of-plane saturation field is 100's of mT [8]. We will treat the MNP as a $+z$ magnetized magnetic dipole located at a height of $d + R_P$ above the disk (as in [11]). This allows us to limit the size of the simulation geometry to a cuboid containing only the disk. Physically, d (10 – 100 nm) represents the separation between the bottom of the spherical particle and the top of the disk (passivation layers and contacts will limit the minimum d in real devices [24]). For simplicity, most simulations are carried out in zero external magnetic field and we artificially set $M_{S,P}$ (the MNP's M_S) to 200 kA/m (the dipole moment is calculated as $\frac{4}{3}\pi R_P^3 M_{S,P}$). Note that a (relatively easy to generate) 200 mT field can induce this moment in magnetically clustered MNPs [27] and this (field-dependent, non-saturated) moment has been successfully used in simulations to reproduce experimentally observed MNP-modified resonances [28]. We will however confirm that comparable results are obtained in a finite OOP field.

For each simulation, we set a vortex-like magnetic state, optionally add a dipole and/or external magnetic field and then compute the equilibrium magnetization configuration in the ferromagnetic disk using MuMax3's internal relaxation routine, 'relax()'. We concentrate first on results where the gyrotropic mode is excited through the application of a transverse field pulse with a sinc function time dependence (frequency cutoff of 30 GHz, offset of 300 ps and amplitude of 2 mT). Such a field induces a displacement of the vortex core which is followed by damped, small amplitude (~ 1 nm) gyrotropic core dynamics (referred to here as 'ringdown' dynamics). We extract f_G from a discrete Fourier transform of the time dependent, x -component of the spatially averaged disk magnetization (total time = 100 ns). In the absence of an applied field, $f_G = f_{G,0} \approx 580$ MHz. We will mostly consider changes in f_G measured relative to $f_{G,0}$ ($\Delta f_G = f_G - f_{G,0}$).

III. PARTICLE-DRIVEN GYROTROPIC FREQUENCY SHIFTING

There are three mechanisms by which a MNP's stray field can modify f_G . Below, we consider how these mechanisms change the core stiffness coefficient, κ , and/or the gyroconstant, G , which together determine f_G via [6]:

$$2\pi f_G = \frac{\kappa}{G}. \quad (1)$$

Fig. 2. (a) Δf_G as a function of in-plane (IP) or out-of-plane (OOP) magnetic field (static and spatially uniform). The thin black line is a linear fit to the OOP data going through (0,0). Insets (i-iii) show maps of the z -component of the equilibrium magnetization configuration (m_z) for the fields marked by the arrows (m_z -range is -0.03 to 1). One can identify the core position (light dot) and/or disk-wide spin canting [disk-wide purple shading, e.g., inset (iii)]. (b,c) Δf_G values induced by IP or OOP field components of a particle with width, $2R_P$, of 100 nm or 500 nm with a disk-particle separation, d , of 10 nm. Data is shown for a particle which is (b) laterally centered or (c) offset by 150 nm in the $+x$ direction. As above, at-equilibrium maps of m_z are shown for each condition: insets (i-iv). The m_z -range here is -0.03 to 0.21. This facilitates visualization of localized canting but makes the vortex core ($m_z = 1$ at its center) appear wider.

κ describes the restoring force acting on the vortex core when the core is displaced from its equilibrium position. For small radial core displacements, X , one can assume a parabolic scaling of the disk's magnetic energy, W : $W(X) = W(0) + \frac{1}{2}\kappa X^2$ (the confinement potential is however typically anharmonic in reality [9]). The main contribution to κ is typically magnetostatic [6], [35] however exchange or Zeeman energies [12], [11] and Oersted fields [36] can be important in certain scenarios. The gyroconstant, G , describes the strength of the lateral force acting on the vortex core due to its motion. It can be calculated from the thickness averaged spin structure using $G = (M_S L / \gamma) \int_A \mathbf{m} \cdot (d\mathbf{m}/dx \times d\mathbf{m}/dy) dx dy$ [37], [38], [6] where $\mathbf{m}$ is the normalized magnetization vector. For small amplitude motion it has been shown that the integration should be carried out over the vortex core [11].

The three mechanisms by which a dipole field can modify f_G (Eq. (1)) are as follows. (i) 'Core confinement' occurs when localized out-of-plane fields aligned with the core magnetization are centered above the vortex core [11]. These fields confine the core, making lateral displacements energetically unfavorable. This increases κ and thus f_G . (ii) 'Spin canting' arises in the presence of spatially (quasi)uniform OOP fields which generate an OOP canting of the vortex's in-plane curling

magnetization [Fig. 2(a), inset iii]. Canting parallel to the core reduces both κ and G [8], [12]. However, reductions in G dominate, resulting in an increased f_G [8], [12]. (iii) ‘Core shifting’ occurs in the presence of IP fields which shift the vortex core within its anharmonic confining potential [9]. In symmetric disks, shifts of the core away from the disk’s center increase κ and thus f_G [10], [9] [Fig. 2(a), inset ii].

In Fig. 2(b) we show the Δf_G induced independently by the IP and OOP components of dipole fields generated by particles with 100 nm or 500 nm diameters centered above the disk with $d = 10$ nm. The OOP field component results in a positive frequency shift for both particle sizes [Figs. 2(b)(i,iii)]. Note that Δf_G is larger for the 100 nm particle despite its smaller volume leading to a weaker stray field. This is because the 100 nm particle’s highly localized field leads to significant core confinement which strongly enhances the Δf_G per unit field when compared to the larger particle’s wide-scale spin canting [11]. Note that due to the laterally centered position of the particle, the IP components of the centered MNPs’ fields are oriented radially inward toward the core and primarily modify the curling magnetization via an inward canting of the spins. For the 100 nm MNP, this generates a weak, negative Δf_G [Fig. 2(b)(ii)].

For a lateral, rightward ($+x$) shift of the MNP position of 150 nm, only the 500 nm MNP induces a clear Δf_G [Fig. 2(c)]. Although the 100 nm MNP generates changes in the curling magnetization, these are localized far from the core position [e.g., Fig. 2(c)(i)] and lead to $\Delta f_G \sim 0$. In contrast, the 500 nm MNP creates a broad stray field distribution, its OOP component generating an OOP spin-canting that is concentrated beneath the particle on the disk’s right hand side. Visible canting does however extend to the core’s location [unlike for the smaller MNP in Fig. 2(c)(i)], leading to an increase in f_G [Fig. 2(c)(iii)]. We also note that due to the MNP’s offset position, the IP component of this MNP’s dipolar field is no longer radially oriented towards the disk’s center and thus can act on the vortex’s curling spin structure to strongly shift the core’s position [Fig. 2(c)(iv)], again increasing f_G .

In Fig. 3(a) we present data for four different particle sizes ($d = 10$ nm) showing how Δf_G depends on the lateral dipole position along a line passing through the center of the disk for full ‘3-dimensional’ dipole fields (i.e. IP and OOP components). Also shown is the particle’s out-of-plane (solid black lines) and in-plane (dashed black lines) fields as calculated at the lateral center of the disk (averaged across the disk’s thickness) and the equilibrium core displacement for each dipole position [Fig. 3(b)]. As seen above, the smallest particles (100 nm and 200 nm widths) generate shifts only when close to the center of the disk with $\Delta f_G \approx 0$ for displacements above $\sim R_p$. For centered dipoles, Δf_G is about equal for the two smallest MNPs despite the 100 nm particle generating a much weaker (but more localized) field, a consequence of core confinement. For the two larger particle sizes, Δf_G is greater than 10 MHz even at the maximum dipole offset of 300 nm with two peaks appearing in the Δf_G versus position curve. Indeed, the comparatively stronger and less localized IP field component of these larger MNPs remains capable of strongly shifting the core equilibrium position (and thus

Fig. 3. (a) Δf_G (red squares) due to the full particle-generated stray field (i.e. both IP and OOP field components) as a function of particle position for a (i) 100 nm, (ii) 200 nm, (iii) 400 nm and (iv) 500 nm MNP. Also shown in these figures is the particle’s out-of-plane (solid black lines) and in-plane (dashed black lines) field as calculated at the disk’s center. (b) The distance of the vortex core from the disk center as a function of particle position for each particle size.

modifying f_G) even for large dipole displacements [10’s of nm in Figs. 3(a,b)(iii,iv)]. We find that the maximum Δf_G arises for dipole positions where the IP field component at the center of the disk is close to maximized. Note that for the large particles that are centered above the disk (i.e. where there is no lateral core displacement and confinement effects are weak due to the large MNPs’ broad stray fields) there is still a significant Δf_G due to spin-canting [Fig. 2(b)(iii)].

An increased disk-particle separation reduces Δf_G [Fig. 4] since the dipolar field acting on the disk is weakened. The percentage reduction in Δf_G per unit of vertical displacement is however clearly strongest for the 100 nm particle since the weaker and broader stray field profile acting on the disk

Fig. 4. (a,b) Δf_G due to the full particle-generated stray field (i.e. both IP and OOP field components) as a function of particle position for three disk-particle separations for (a) 100 nm and (b) 400 nm particles. The inset to (a) plots the out-of-plane stray field at the disk center (averaged across the disk thickness) as a 100 nm particle is shifted laterally across the disk at a height of $d = 10$ nm and $d = 50$ nm (y -axis ranges from -5 mT to 65 mT, x -axis ranges from -200 nm to 200 nm).

for increased d [Fig. 4(a) inset] significantly reduces core-confinement [11]. We do however observe a qualitatively similar particle-position- and particle-size-dependence of Δf_G : a single peak for the 100 nm MNP and a double peak for the 400 nm MNP. Note however that for the 400 nm particle, the shoulders in the Δf_G profile (linked to core shifting) become less distinct as d is increased, consistent with a decreased core displacement due to a weaker stray field. We finally note that the simulated Δf_G values for the 400 nm particle well exceed typically measured vortex nano-oscillator linewidths (down to ~ 1 MHz or less) [17], [18], [39], [40] which is encouraging for electronic detection (this is also true for the smaller particles, but with stricter restrictions on the particle position).

IV. LARGE AMPLITUDE CORE DYNAMICS

Instead of small amplitude ringdown dynamics, we now consider larger (and more experimentally relevant; e.g., [41]) core orbits. Here we apply a transverse, radiofrequency magnetic field with amplitude 0.8 mT and frequency close to the f_G determined from the ringdown simulations. This induces gyrotropic core motion (steady state orbit radius ≈ 50 nm with gyration around the core's equilibrium position) which we refer to as ‘driven’ dynamics. The driving frequency was stepped and that which induced the largest orbit radius after 50 ns was taken to be f_G (a 5 MHz step size yielded an intrinsic uncertainty in the extracted f_G of ± 2.5 MHz).

Δf_G values as a function of particle position for ringdown (red squares) and driven (blue diamonds) gyrotropic dynamics are compared in Figs. 5(a)-(c). When the particle is placed at the disk center, there is a clear reduction in Δf_G for large amplitude, driven oscillations of the vortex core. This reduction is most significant for small particle sizes [simulated down to $2R_P = 40$ nm; Fig. 5(a)] with Δf_G falling by $\sim 88\%$ for the 40 nm MNP compared to $\sim 23\%$ for the 400 nm MNP. The strong reduction in Δf_G for small MNPs and driven dynamics is consistent with the vortex core trajectory now being outside of the strongest part of the MNP's localized out-of-plane stray field. This weakens localized-field-induced confinement effects, decreasing Δf_G . As the MNP size is increased however, the reduction in Δf_G becomes less significant since the Δf_G for large particles is due to wide-scale spin-canting across the disk which will be present throughout the core orbit. Changes in the gyrotropic frequency due to MNP-induced shifts in the core's equilibrium position are also still present for driven dynamics. As such, the characteristic double peak in Δf_G for large MNPs is observed for both ringdown and driven dynamics [Fig. 5(c)].

Finally, we show Δf_G (determined from ringdown dynamics) as a function of particle position for a 100 nm and 400 nm MNP in the presence of a spatially uniform 200 mT out-of-plane field [Fig. 5(d)]. We observe a similar particle size dependence of Δf_G [now measured with respect to f_G in a 200 mT OOP field; see Fig. 2(a)]. Our results appear to be qualitatively independent of the presence of an out-of-plane magnetic field as long as that field is not so large as to destroy the vortex state or lead to a deviation in the linear OOP field dependence of f_G [12].

Fig. 5. (a-c) Δf_G due to the full particle-generated stray field (i.e. both IP and OOP field components) as a function of particle position for sinc-field-driven ringdown dynamics (red squares) and sinusoidal-field-driven dynamics (blue diamonds) for a (a) 40 nm (b) 100 nm and (c) 400 nm particle. The disk-particle separation is $d = 10$ nm. (d) Δf_G as a function of particle position in the presence of a spatially uniform 200 mT out-of-plane field (we neglect any influence of stray fields generated by the disk's canted magnetization on the particle moment). This is shown for a 100 nm diameter particle (blue squares) and a 400 nm diameter particle (red circles).

V. CONCLUSION

In summary, both the IP and OOP components of MNP-generated stray fields can significantly modify the gyrotropic vortex resonance frequency, f_G , in a ferromagnetic disk ($\Delta f_G \gtrsim 10$'s of MHz). Although small particles (≤ 200 nm in width in this study) can produce significant changes in f_G via core confinement, these changes are strongly reduced when the core trajectory is not localized directly beneath the particle. In contrast, larger particles (≥ 400 nm) can create strong changes in f_G even when they are 300 nm from the vortex core. This is because these particles create broad, strong stray field profiles which modify the gyrotropic frequency via wide-scale canting of the vortex's curling spins and significant shifts of the core within its confining potential. The observation of a large Δf_G due to these latter frequency shifting mechanisms is less dependent on particle placement and core orbit size. As such, the use of larger particles likely represents a more reliable sensing approach, especially if particle position cannot be precisely controlled.

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