Dynamic Scaling of Ferromagnetic Micro-rod Clusters under a Weak Magnetic Field

Rui Cheng¹, Lu Zhu¹, Weijie Huang², Leidong Mao¹ and Yiping Zhao²

¹College of Engineering, University of Georgia, Athens, Georgia 30602, USA

²Department of Physics and Astronomy, University of Georgia, Athens, Georgia 30602, USA

Supporting Information

S1. FMR Characterization

The crystal structures of as synthesized FMRs were characterized by an X-ray diffractometer (XRD; PANalytical X'Pert PRO MRD) with a Cu K α source (λ = 1.5405980 Å) at 45 kV and 40 Ma (Figure S1a). The diffraction angle scanning range was from 15° to 70° at an angular step of 0.01°. Magnetic properties of as-synthesized Fe₃O₄ microrods were measured at room temperature by using a Vibrating Sample Magnetometer (VSM, Model EZ7; MicroSense, LLC, Lowell, MA, USA) with a 2.15 T electromagnet (Figure 1b). The magnetic moment of the sample was measured over a range of applied fields from -1.5 to +1.5 kOe. The measurements were conducted in step field mode at a step size of 250 Oe s⁻¹. The statistic dimensions of assynthesized FMRs (Figure S1b, S1c and S1d) and the structures of FMRCs (Figure 2c and 2d) were characterized by a field-emission scanning electron microscope (FESEM) equipped with an energy dispersive X-ray spectroscopy (FEI Inspect F).

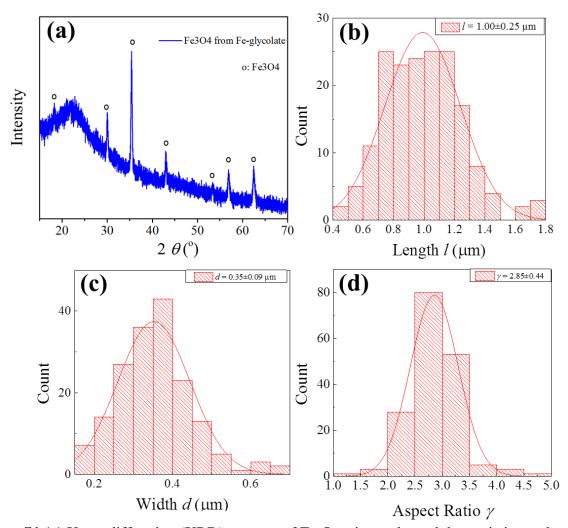


Figure S1 (a) X-ray diffraction (XRD) patterns of Fe₃O₄ microrods; and the statistic results of FMRs regarding to (b) length l, (c) width d and (d) aspect ratio γ .



Figure S2 Smaller particles formed during the annealing process which may cause a smaller measured M_r/M_s value.

S2. Experimental Data of FMRCs' Dynamics under Different B field

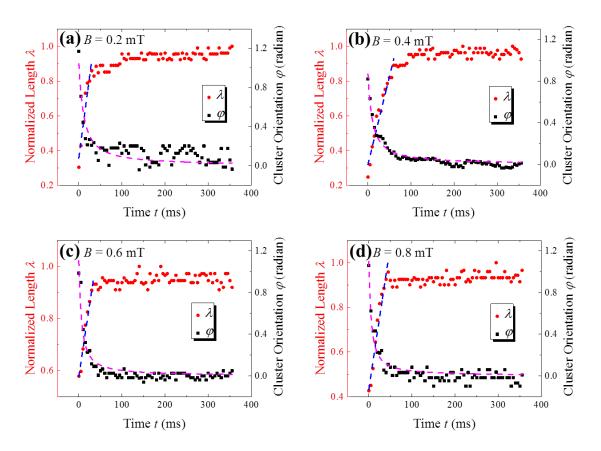


Figure S3 Representative plots of the normalized length λ (red circle) and the orientation φ (black square) with respect to time t at (a) B = 0.2 mT, (b) B = 0.4 mT, (c) B = 0.6 mT and (d) B = 0.8 mT. All the dash curves are the fitting results by Eq. (10).

S3. Derivation of Dynamic Equations of FMRC Consisting of 2N and 2N+1 FMRs

All the following derivation is based on the sketch shown in Figure 3b.

Let's start from Eqs. (6) and (7)

$$2mB\sin\varphi\cos\theta = \left(\sum_{k=p+2}^{N} F_{D_{\perp}}^{k} + 2\sum_{k=p+1}^{N} F_{D_{\perp}}^{k} + \sum_{k=p}^{N} F_{D_{\perp}}^{k}\right) \frac{l}{2}\cos\theta - \left(F_{n}^{(p+1)+} - F_{n}^{p-}\right) \frac{l}{2}\sin\theta, \tag{6}$$

$$2mB\cos\varphi\sin\theta = \left(\sum_{k=p}^{N} F_{D_{\parallel}}^{k} + 2\sum_{k=p+1}^{N} F_{D_{\parallel}}^{k} + \sum_{k=p+2}^{N} F_{D_{\parallel}}^{k}\right) \frac{l}{2}\sin\theta - \left(F_{f}^{(p+1)+} - F_{f}^{p-}\right) \frac{l}{2}\cos\theta. \tag{7}$$

For 2N+1 FMRs model, the cluster has one single rod at its center and N rods on both of its sides evenly. Due to the symmetry of the system, we only need consider the right half of the cluster. If N is an odd number, we need include the central rod (denoted as 0^{th} rod) in the right part of the cluster in order to apply Eqs. (6) and (7), which are based on the unit pair of rods.

$$(N+1)mB\sin\varphi\cos\theta = \sum_{p=1}^{N} \sum_{k=p}^{N} F_{D_{\perp}}^{k} l\cos\theta + F_{n}^{0-} \frac{l}{2}\sin\theta, \qquad (S1-1)$$

$$(N+1)mB\cos\varphi\sin\theta = \sum_{n=1}^{N} \sum_{k=n}^{N} F_{D_{\parallel}}^{k} l\sin\theta + F_{f}^{0-} \frac{l}{2}\cos\theta.$$
 (S1-2)

Applying the same mathematical treatment on the left part of the cluster (including the central rod as well), we get the exactly the same expressions of Eqs. (S1-1) and (S1-2). Since the torques on the central rod are employed twice for the calculation, we need to remove them one time when we added the torque relations of the both sides to estimate the relations on the entire cluster

$$2(N+1)mB\sin\varphi\cos\theta - mB\sin(\varphi+\theta) = 2\sum_{p=1}^{N}\sum_{k=p}^{N}F_{D_{\perp}}^{k}l\cos\theta - F_{f}^{0-}l\cos\theta, \qquad (S2-1)$$

$$2(N+1)mB\cos\varphi\sin\theta - mB\sin(\varphi+\theta) = 2\sum_{p=1}^{N}\sum_{k=p}^{N}F_{D_{\parallel}}^{k}l\sin\theta - F_{n}^{0-}l\sin\theta.$$
 (S2-2)

Plug in all the expressions of the forces (i.e., $F_{D_{\parallel}}^{p} = -\eta \varepsilon_{\parallel} p l \dot{\theta} \sin \theta$ and $\vec{F}_{D_{\perp}}^{p} = -\eta \varepsilon_{\perp} p l \dot{\phi} \cos \theta$) in Eqs. (S2-1) and (S2-2)

$$mB\sin\varphi\cos\theta - \frac{mB}{2N}\cos\varphi\sin\theta = \left(-\frac{N^2}{3} - \frac{N}{4} + \frac{1}{12}\right)\eta\varepsilon_{\perp}\dot{\varphi}l^2\cos^2\theta, \qquad (S3-1)$$

$$mB\cos\varphi\sin\theta - \frac{mB}{2N}\sin\varphi\cos\theta = \left(-\frac{N^2}{3} - \frac{N}{4} + \frac{1}{12}\right)\eta\varepsilon_{\parallel}\dot{\theta}l^2\sin^2\theta. \tag{S3-2}$$

When N is big enough (e.g., N > 5), the terms regarding to N with lower exponents can be ignored in the both sides of Eqs. (S3-1) and (S3-2), then we have

$$\dot{\varphi} = -\frac{3mB\sin\varphi}{\varepsilon_{\perp}\eta N^2 l^2\cos\theta},\tag{S4-1}$$

$$\dot{\theta} = -\frac{3mB\cos\varphi}{\varepsilon_{\parallel}\eta N^2 l^2 \sin\theta} \,. \tag{S4-2}$$

Obviously, Eqs. (S4-1) and (S4-2) are the same to Eq. (8).

If N is an even number, we need exclude the central rod (denoted as 0^{th} rod) in the right part of the cluster and apply Eqs. (6) and (7) on the right cluster

$$NmB\sin\varphi\cos\theta = \sum_{p=2}^{N} \sum_{k=p}^{N} F_{D_{\perp}}^{k} l\cos\theta + \sum_{k=1}^{N} F_{D_{\perp}}^{k} \frac{1}{2}\cos\theta + F_{n}^{1-} \frac{1}{2}\sin\theta,$$
 (S5-1)

$$NmB\cos\varphi\sin\theta = \sum_{p=2}^{N} \sum_{k=p}^{N} F_{D_{||}}^{k} l\sin\theta + \sum_{k=1}^{N} F_{D_{||}}^{k} \frac{l}{2} \sin\theta + F_{f}^{1-} \frac{l}{2} \cos\theta.$$
 (S5-2)

Due to the symmetry of the system, the summation of rod pairs over the whole cluster equals to twice of Eqs. (S5-1) and (S5-2) with the complement of the central rod

$$2NmB\sin\varphi\cos\theta + mB\sin(\varphi-\theta) = 2\sum_{p=2}^{N}\sum_{k=p}^{N}F_{D_{\perp}}^{k}l\cos\theta + 2F_{f}^{1-}l\cos\theta, \qquad (S6-1)$$

$$2NmB\cos\varphi\sin\theta + mB\sin(\varphi-\theta) = 2\sum_{p=2}^{N}\sum_{k=p}^{N}F_{D_{\parallel}}^{k}l\sin\theta + 2F_{f}^{1-}l\cos\theta.$$
 (S6-2)

Plug in all the expressions of the forces in Eqs. (S6-1) and (S6-2), and ignore the terms with respect to the lower-order of N, we will obtain the same expressions of Eqs. (S4-1) and (S4-2), i.e., Eq (8). For 2N FMRs model, the cluster has its center on the joint of the two central rods with N rods on its both sides evenly. If N is an odd number, we exclude the first rod (denoted as 1^{st} rod) in the right part of the cluster and then sum Eqs. (6) and (7) of the rest pairs of the rods

$$(N-1)mB\sin\varphi\cos\theta = \sum_{p=2}^{N} \sum_{k=p}^{N} F_{D_{\perp}}^{k} l\cos\theta - \sum_{k=2}^{N} F_{D_{\perp}}^{k} \frac{l}{2}\cos\theta + F_{n}^{2-} \frac{l}{2}\sin\theta,$$
 (S7-1)

$$(N-1)mB\cos\varphi\sin\theta = \sum_{p=2}^{N}\sum_{k=p}^{N}F_{D_{\parallel}}^{k}l\sin\theta - \sum_{k=2}^{N}F_{D_{\parallel}}^{k}\frac{l}{2}\sin\theta + F_{f}^{2-}\frac{l}{2}\cos\theta.$$
 (S7-2)

Similarly, the summation on the whole cluster is the double of Eqs. (S7-1) and (S7-2), adding the corresponding relations of the torques on the central pair of the rods

$$2NmB\sin\varphi\cos\theta = 2\sum_{p=2}^{N}\sum_{k=p}^{N}F_{D_{\perp}}^{k}l\cos\theta + \sum_{k=1}^{N}F_{D_{\perp}}^{k}l\cos\theta + \sum_{k=2}^{N}F_{D_{\parallel}}^{k}l\sin\theta,$$
 (S8-1)

$$2NmB\cos\varphi\sin\theta = 2\sum_{p=2}^{N}\sum_{k=p}^{N}F_{D_{\parallel}}^{k}l\sin\theta + \sum_{k=2}^{N}F_{D_{\perp}}^{k}l\cos\theta + \sum_{k=1}^{N}F_{D_{\parallel}}^{k}l\sin\theta.$$
 (S8-2)

Notice that the expressions of the forces are different from 2N+1 FMRs model. Here, we have

$$F_{D_{\parallel}}^{\ p} = -\eta \varepsilon_{\parallel} \bigg(p - \frac{1}{2} \bigg) l \dot{\theta} \sin \theta \ \ \text{and} \ \ \vec{F}_{D_{\perp}}^{\ p} = -\eta \varepsilon_{\perp} \bigg(p - \frac{1}{2} \bigg) l \dot{\phi} \cos \theta \ . \ \text{Plug the expressions of the forces in}$$

Eqs. (S8-1) and (S8-2)

$$mB\sin\varphi\cos\theta = -\left(\frac{N^2}{3} - \frac{1}{12}\right)\eta\varepsilon_{\perp}\dot{\varphi}l^2\cos^2\theta - \left(\frac{N}{4} - \frac{1}{4N}\right)\eta\varepsilon_{\parallel}\dot{\theta}l^2\sin^2\theta, \quad (S9-1)$$

$$mB\cos\varphi\sin\theta = -\left(\frac{N^2}{3} - \frac{1}{12}\right)\eta\varepsilon_{\parallel}\dot{\theta}l^2\sin^2\theta - \left(\frac{N}{4} - \frac{1}{4N}\right)\eta\varepsilon_{\perp}\dot{\varphi}l^2\cos^2\theta.$$
 (S9-2)

Ignore the terms with respect to the lower-order N, we will obtain the same expressions of Eqs. (S4-1) and (S4-2). If N is an even number, we directly apply the Eqs (6) and (7) on the entire cluster,

$$2NmB\sin\varphi\cos\theta = 2\sum_{p=2}^{N}\sum_{k=p}^{N}F_{D_{\perp}}^{k}l\cos\theta + \sum_{k=1}^{N}F_{D_{\perp}}^{k}l\cos\theta + \sum_{k=1}^{N}F_{D_{\parallel}}^{k}l\sin\theta,$$
 (S10-1)

$$2NmB\cos\varphi\sin\theta = 2\sum_{p=2}^{N}\sum_{k=p}^{N}F_{D_{||}}^{k}l\sin\theta + \sum_{k=1}^{N}F_{D_{||}}^{k}l\sin\theta + \sum_{k=1}^{N}F_{D_{||}}^{k}l\cos\theta.$$
 (S10-2)

Comparing Eqs. (S10-1) and (S10-2) with Eqs. (S8-1) and (S8-2), we should obtain the same results after the simplification.

S4. Derivation for the Analytical Solution of Eq. (9)

Let's $\alpha = \frac{1}{\tau}$, and we separate Eq. (9) into two equations

$$\dot{\lambda} = \alpha \cos \varphi, \tag{S11-1}$$

$$\dot{\varphi} = -\frac{\alpha \sin \varphi}{\lambda} \,. \tag{S11-2}$$

Take the derivation with respect to time *t* for Eq. (S11-1)

$$\ddot{\lambda} = -\alpha \dot{\varphi} \sin \varphi \,. \tag{S12}$$

Replace $\dot{\varphi}$ in Eq. (S12) with its expression in Eq. (S11-2)

$$\ddot{\lambda} = \alpha^2 \frac{\sin^2 \varphi}{\lambda} \to \ddot{\lambda} \lambda = \alpha^2 \sin^2 \varphi . \tag{S13}$$

From Eq. (S11-1), we also have

$$\dot{\lambda}^2 = \alpha^2 \cos^2 \varphi \,. \tag{S14}$$

Add Eq. (S13) to Eq. (S14)

$$\ddot{\lambda}\lambda + \dot{\lambda}^2 = \alpha^2. \tag{S15}$$

Eq. (S15) is an ODE for λ and t.

In addition, take the derivation with respect to time t for Eq. (S11-2)

$$\ddot{\varphi} = -\alpha \left(\frac{\dot{\varphi}\cos\varphi}{\lambda} - \frac{\dot{\lambda}\sin\varphi}{\lambda^2} \right). \tag{S16}$$

Replace $\dot{\lambda}$ and $\dot{\phi}$ in Eq. (S16) with Eqs. (S11-1) and (S11-2),

$$\ddot{\varphi} = \frac{\alpha^2}{\lambda^2} \sin 2\varphi \,. \tag{S17}$$

From Eq. (S11-2), we also have $\frac{\alpha}{\lambda} = \frac{-\dot{\varphi}}{\sin{\varphi}}$. Plug it into Eq. (S17),

$$\ddot{\varphi} = 2\dot{\varphi}^2 \cot \varphi \,. \tag{S18}$$

Eq. (S18) is an ODE for φ and t. Thus, we have decoupled Eq. (9) into Eqs. (S15) and (S18).

To solve Eq. (S15), let $\mu = \lambda^2$. Take the second order derivation with respect to t for μ , we have,

$$\ddot{\mu} = 2\dot{\lambda}\dot{\lambda} + 2\lambda\ddot{\lambda} = 2\left(\dot{\lambda}^2 + \lambda\ddot{\lambda}\right). \tag{S19}$$

Then, Eq. (S15) becomes,

$$\ddot{\mu} = 2\alpha^2. \tag{S20}$$

Solve Eq. (S20),

$$\mu = \alpha^2 t^2 + C_1 t + C_2. \tag{S21}$$

Since $\lambda \approx \cos\theta$, then $0 \le \mu \le 1$. We have a constrained condition,

$$\alpha^2 t^2 + C_1 t + C_2 - 1 \le 0. (S22)$$

However, since α^2 is always positive, so Eq.(S22) cannot be satisfied by all t. Then, λ is expressed as

$$\lambda(t) = \sqrt{\alpha^2 t^2 + C_1 t + C_2}, \lambda(t) \le 1.$$
 (S23)

 $\lambda(t)$ must be positive for its physical meaning. The constant C_2 can be determined by Eq. (S23) at t=0

$$\lambda(t=0) = \sqrt{C_2} \to C_2 = \lambda_0^2, \tag{S24}$$

and the constant C_1 can be determined by taking the first derivation of Eq.(S23) with respect to t

$$\dot{\lambda}(t=0) = \frac{\frac{1}{2}(2\alpha^2t + C_1)}{\sqrt{\alpha^2t^2 + C_1t + C_2}}\bigg|_{t=0} = \frac{C_1}{2\sqrt{C_2}}.$$
 (S25)

According to Eq. (S11-1), we have

$$\dot{\lambda}(t=0) = \alpha \cos \varphi_0 \,, \tag{S26}$$

and then

$$\frac{C_1}{2\sqrt{C_2}} = \alpha \cos \varphi_0 \to C_1 = 2\alpha \lambda_0 \cos \varphi_0. \tag{S27}$$

Replace α with $\frac{1}{\tau}$ and plug into the expression of C_1 , C_2 in Eq. (S23)

$$\lambda(t) = \sqrt{\left(\frac{t}{\tau} + \lambda_0 \cos \varphi_0\right)^2 + {\lambda_0}^2 \sin^2 \varphi_0}, \lambda(t) \le 1$$
 (S28)

To solve Eq. (S18), we divide it by $\dot{\phi}$

$$\frac{\ddot{\varphi}}{\dot{\varphi}} = 2\dot{\varphi}\cot\varphi \,. \tag{S29}$$

The left side of Eq. (S29) can be rewritten as

$$\frac{\ddot{\varphi}}{\dot{\varphi}} = \frac{d\left(\ln \dot{\varphi}\right)}{dt}.$$
 (S30)

Combine Eqs. (S29) and (S30)

$$\frac{d\left(\ln\dot{\varphi}\right)}{dt} = 2\frac{d\varphi}{dt}\cot\varphi. \tag{S31}$$

Multiply Eq. (S31) with dt

$$d(\ln \dot{\varphi}) = 2\cot \varphi d\varphi. \tag{S32}$$

Rewrite Eq. (S32) in integral form

$$\int_{\varphi_0}^{\varphi} d\left(\ln \dot{\varphi}\right) = \int_{\varphi_0}^{\varphi} 2\cot \varphi d\varphi \to \ln \dot{\varphi} - \ln \dot{\varphi}_0 = 2\left(\ln\left(\sin \varphi\right) - \ln\left(\sin \varphi_0\right)\right). \quad (S33)$$

Then, Eq. (S33) becomes a new ODE

$$\ln \frac{\dot{\varphi}}{\dot{\varphi}_0} = \ln \left(\frac{\sin \varphi}{\sin \varphi_0} \right)^2 \rightarrow \dot{\varphi} = C \sin^2 \varphi, C = \frac{\dot{\varphi}_0}{\sin^2 \varphi_0}.$$
 (S34)

Separate variables of Eq. (S34)

$$\dot{\varphi} = C \sin^2 \varphi \to \frac{d\varphi}{\sin^2 \varphi} = Cdt . \tag{S35}$$

Solve Eq. (S35) by integrating its both sides

$$\int_{\varphi_0}^{\varphi} \frac{1}{\sin^2 \varphi} d\varphi = \int_0^t C dt \to \cot \varphi_0 - \cot \varphi = Ct.$$
 (S36)

Reorganize Eq. (S36)

$$\varphi = \cot^{-1}(C_3 t + C_4), C_3 = -C = \frac{-\dot{\varphi}_0}{\sin^2{\varphi}_0}, C_4 = \cot{\varphi}_0.$$
 (S37)

Plug the expression of $\dot{\varphi}$ in Eq. (S11-2) at t = 0 into C_3

$$C_3 = \frac{-\left(-\alpha \frac{\sin \varphi_0}{\lambda_0}\right)}{\sin^2 \varphi_0} = \frac{\alpha}{\lambda_0 \sin \varphi_0}.$$
 (S38)

Replace replacing α with $\frac{1}{\tau}$ and plug in the expression of C_3 , C_4 in Eq. (S37)

$$\varphi = \cot^{-1} \left(\frac{1}{\lambda_0 \sin \varphi_0} \left(\frac{t}{\tau} \right) + \cot \varphi_0 \right)$$
 (S39)