

Controlling directionality in dipolar colloids via relaxation in alternating rotating fields

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Time-varying external fields can program dipolar interactions in soft matter systems. Continuously rotating fields are widely used to promote isotropic self-assembly, but finite relaxation of an induced dipole generically introduces a phase lag that can drive the spinning of both individual particles and clusters. Alternating rotating fields, which reverse the rotation direction every half cycle, suppress net spinning, but need not yield a truly isotropic time-averaged interaction when the dipole relaxes with a finite timescale τ . Here, we propose an extended alternating rotating field in which the field sweeps beyond 2π within each half cycle to compensate for relaxation-induced undersampling of dipole orientations near the reversal direction. Using paramagnetic colloids under time-varying magnetic fields as a representative realization of a generic first-order relaxation model, we determine the half-cycle sweep angle ϕ_{iso} required to obtain an isotropic cycle-averaged pair potential. The isotropy condition collapses onto a master curve controlled by the dimensionless parameter $\tau\omega$, and we derive accurate analytical approximations in the experimentally important small- $\tau\omega$ regime. We also analyze the residual anisotropy under conventional alternating rotating-field protocols and show that its nonmonotonic dependence on $\tau\omega$ underlies the single-peaked $\phi_{\text{iso}}(\tau\omega)$. Because the theory assumes only first-order relaxation, the framework applies broadly to field-responsive soft matter beyond magnetic colloids. It enables switching between isotropic and anisotropic cycle-averaged pair interactions while suppressing net rotation and provides a route to infer relaxation times.

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I. INTRODUCTION

Field-driven dipolar colloids provide a versatile platform for programming interparticle interactions and studying nonequilibrium self-assembly [1]. By applying electric or magnetic fields, the strength, symmetry, and anisotropy of the effective pair interactions can be tuned dynamically [2–5]. Time-varying fields are especially useful because the cycle-averaged dipole-dipole interaction can differ substantially from the instantaneous interaction, allowing external fields to prescribe effective interaction landscapes for aggregation, crystallization, and model condensed-matter behavior [6–16].

For induced-dipole particles, the equilibrium dipole moment is often approximated as proportional to the applied field. However, under a time-varying field, the induced moment may not reach this equilibrium value instantaneously. Instead, it relaxes toward the field-dependent equilibrium dipole with a characteristic time τ [17,18]. Because the microscopic origin of this delay is not essential for our purposes, we treat τ phenomenologically as a generic first-order relaxation time. It may reflect intrinsic relaxation processes of the induced moment and/or the microstructure of the building blocks, as well as effective delays in the experimental setup

used to generate the external field, such as coil inductance. Such a finite response time matters most under time-varying fields, where the dipole moment acquires a phase lag relative to the field and the resulting time-averaged interactions depend sensitively on τ [19].

As a representative model system, we focus on superparamagnetic micron-sized beads subjected to time-varying magnetic fields. These colloids are multicore magnetic microparticles that consist of a large ensemble of randomly oriented iron-oxide ferromagnetic nanoparticles embedded within a nonmagnetic material [20,21]. Although the magnetic moments of the immobilized nanoparticles relax by the Néel mechanism [22,23], the net induced dipole of a colloidal bead under a time-varying magnetic field can exhibit an out-of-phase component that produces a magnetic torque because of nontrivial relaxation processes [24–27]. For this reason, the dipolar response can deviate from the instantaneous-field approximation at field frequencies of tens to hundreds of hertz. Reported relaxation times for the induced dipole of these beads are on the order of milliseconds—5–8 orders of magnitude longer than those of the individual nanoparticles—highlighting a significant slowdown of the internal magnetic relaxation dynamics [28–33]. Despite this complexity, the overall magnetization of superparamagnetic beads can often be captured by a first-order relaxation equation with a finite effective magnetic relaxation time τ , which reproduces experimental observations of these colloids in time-varying fields [33].

Among time-varying magnetic fields, continuously rotating magnetic fields (CRM) have been widely used to drive self-assembly [12,34], because they can readily render the

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cycle-averaged dipole-dipole interaction isotropic in the plane of rotation [35]. However, for finite τ , the induced dipoles acquire a phase lag in CRMF, which can drive persistent rotation of both single particles and clusters in experiments [36–38]. Similar cluster rotation appears when the building blocks are ferromagnetic colloids with a permanent dipole [39–41], as well as other chiral colloidal systems [42]. This rotation generates a shearlike motion that is absent in atomic condensed-matter systems. Although such behaviors are interesting in their own right, they complicate comparisons with quasistatic atomic condensed states and can introduce additional nonconservative transverse forces [43] that can obscure the interpretation of interaction-driven aggregation.

Alternating rotating magnetic fields (ARMF), in which the sense of rotation is reversed after each half cycle, suppress net rotation. Most previous studies of ARMF have focused on regimes where relaxation effects were expected to be small, or they did not systematically examine relaxation under the experimental conditions considered [2,10,13,44,45]. However, recent work has shown that finite relaxation can bias the induced magnetization over one cycle and render the cycle-averaged dipolar interaction anisotropic [46]. Thus, even though ARMF suppresses net spinning, it does not necessarily produce an isotropic time-averaged interaction. This raises two related questions: When can relaxation effects be neglected, and when they cannot, how can isotropy be restored without sacrificing the no-net-rotation advantage of ARMF?

In this work, we develop a theoretical framework for induced-dipole systems with a first-order relaxation time under an extended alternating rotating magnetic field (EARMF), in which the sweep angle per half cycle exceeds 2π . The additional angular sweep compensates for the relaxation-induced undersampling of dipole orientations near the field-reversal direction. Although the formation and evolution of trimers and larger aggregates are important for understanding isotropic self-assembly, they also involve assembly kinetics and many-body effects. We therefore focus on the pair-level condition for orientational isotropy, which provides a microscopic basis for designing isotropic self-assembly. Starting from a first-order relaxation equation, we derive analytical expressions for the steady-state dipole dynamics and the cycle-averaged pair potential. We show that the isotropy condition collapses onto a single master curve governed by the dimensionless parameter $\tau\omega$, where ω is the external-field angular frequency. We also provide accurate analytical approximations in the experimentally relevant small- $\tau\omega$ regime. Together, these results provide quantitative pair-level design rules for controlling cycle-averaged interparticle interactions under EARMF protocols through the sweep angle and driving frequency.

II. THEORETICAL RESULTS

If the relaxation time is finite, the magnetization cannot align with the external field instantaneously [Fig. 1(a)]. As a result, even under a regular ARMF (with half-cycle sweep angle $\phi = 2\pi$), where the field direction uniformly samples the xy plane over one cycle, the lagging magnetization cannot fully reach the directions near the switching axis (the x axis in our coordinate system) before the field rotation is inverted. The directions near the x axis are therefore undersampled by

the magnetization vector, and the cycle-averaged magnetization distribution becomes concentrated away from the x axis. This anisotropy in the magnetization distribution produces an anisotropic component in the cycle-averaged dipole-dipole interactions. Consequently, the dimer preferentially aligns perpendicular to the switching axis of the field rotation, i.e., along the y axis [Fig. 1(b)]. This observation motivates an extended protocol in which the field sweeps beyond 2π during each half cycle to compensate for the relaxation-induced deficit near the switching axis. We refer to this protocol with $\phi > 2\pi$ as the EARMF. In this section, we analytically evaluate the cycle-averaged pair potential $\langle U_{ij} \rangle$ as a function of the half-cycle sweep angle ϕ and determine the isotropic sweep angle ϕ_{iso} at which the orientation-dependent component vanishes.

First, we define the EARMF protocol shown in Fig. 1(c). In the first half cycle, the field rotates counterclockwise (CCW) from $\psi = -\alpha$ to $\psi = +\alpha$, completing one full turn in between; the total swept angle is $\phi = 2\pi + 2\alpha$. In the second half cycle, the field reverses and rotates clockwise (CW) from $+\alpha$ back to $-\alpha$, again completing one full turn and sweeping the same angle ϕ . In both half cycles, the in-plane field angle $\psi(t)$ changes with a constant angular speed, $\omega = |d\psi/dt|$. The protocol is periodic with period $T_{\text{cyc}} = 2\phi/\omega$, with a reversal of the rotation direction every half cycle. For convenience, we introduce the half-cycle index $N_h = \lfloor t/(T_{\text{cyc}}/2) \rfloor$ and the reduced time $t' = t - N_h(T_{\text{cyc}}/2) \in [0, T_{\text{cyc}}/2)$, where even (odd) N_h corresponds to CCW (CW) rotation. The applied magnetic field is written as

$$\begin{aligned} \mathbf{H}_0(t) &= H_0[\cos \psi(t) \hat{\mathbf{e}}_x + \sin \psi(t) \hat{\mathbf{e}}_y], \\ \psi(t) &= (-1)^{N_h}(-\alpha + \omega t'). \end{aligned} \quad (1)$$

In the linear magnetization regime, far from magnetic saturation, the induced magnetic dipole moment of the i th particle, $\mathbf{m}_i(t)$, can be modeled as [17,46]

$$\frac{d\mathbf{m}_i(t)}{dt} = -\frac{1}{\tau}[\mathbf{m}_i(t) - \chi_p(\mathbf{H}_0(t) + \mathbf{H}_{L,i}(t))] + \boldsymbol{\Omega}_i(t) \times \mathbf{m}_i(t). \quad (2)$$

This equation describes relaxation toward the instantaneous effective field $\mathbf{H}_0 + \mathbf{H}_{L,i}$, together with the rotation of the magnetization vector caused by the rigid-body rotation of the particle. Here, τ is the effective relaxation time of the particle, and $\chi_p = (4\pi a^3/3)\chi$ is the particle susceptibility, with the particle radius a and the intrinsic susceptibility χ . The vector $\boldsymbol{\Omega}_i$ denotes the angular velocity associated with the particle's own rotation, and the term $\boldsymbol{\Omega}_i \times \mathbf{m}_i$ accounts for this rotational contribution. The interaction field acting on particle i , denoted by $\mathbf{H}_{L,i}(t)$, is the sum of the dipolar fields produced by all neighboring induced dipoles and is given by

$$\mathbf{H}_{L,i}(t) = \sum_{j \neq i} \frac{1}{4\pi r_{ij}^3} [3(\mathbf{m}_j(t) \cdot \hat{\mathbf{r}}_{ij})\hat{\mathbf{r}}_{ij} - \mathbf{m}_j(t)], \quad (3)$$

where $\mathbf{r}_{ij}$ is the separation vector from particle j to i and $\hat{\mathbf{r}}_{ij} = \mathbf{r}_{ij}/r_{ij}$.

As a first approximation, we neglect both the interaction field $\mathbf{H}_{L,i}$ and the rotational term $\boldsymbol{\Omega}_i \times \mathbf{m}_i$, reducing Eq. (2) to a single-particle linear relaxation equation. This simplified

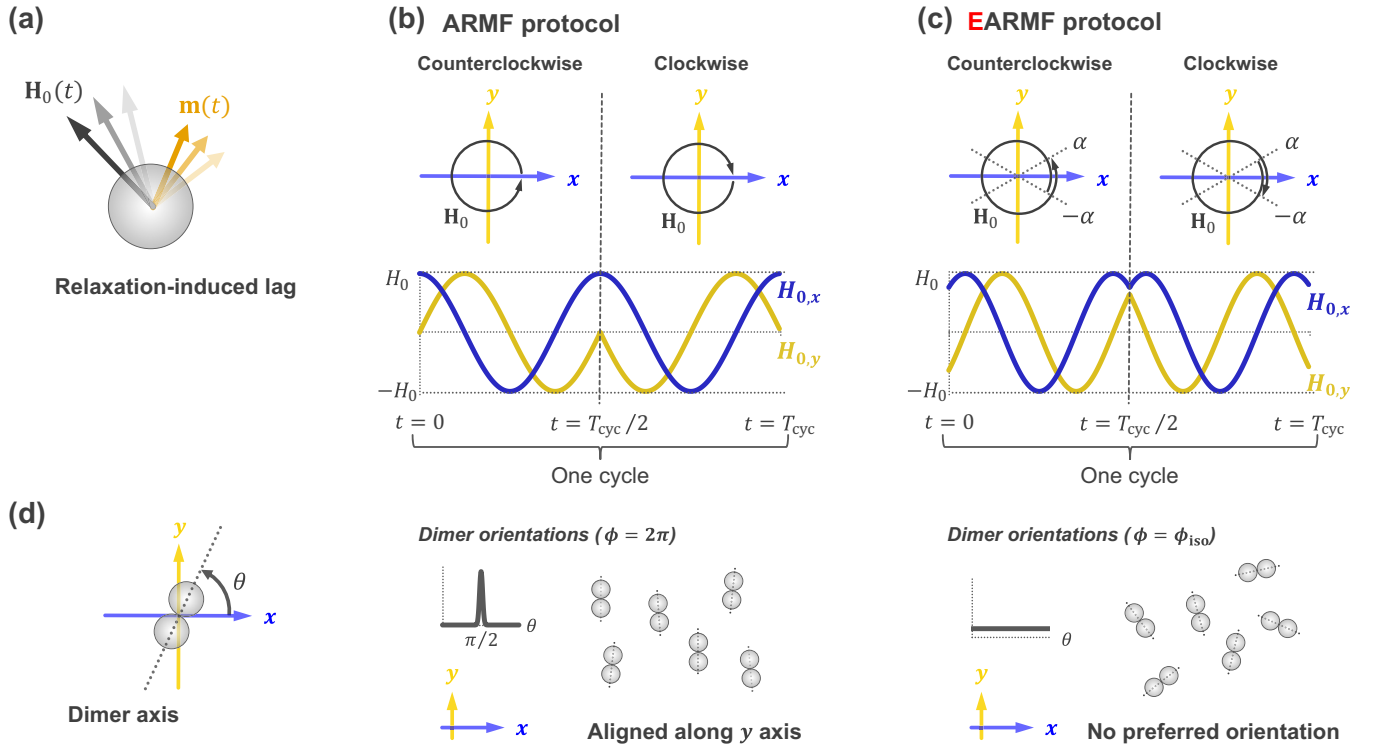


FIG. 1. (a) Schematic illustration of relaxation-induced magnetization lag for a single superparamagnetic particle under a time-varying magnetic field (arbitrary scale). (b) Regular ARMF protocol and schematic illustration of the resulting orientational selectivity predicted from the cycle-averaged pair potential. Top: $\mathbf{H}_0(t)$ rotates at angular speed ω and sweeps a half-cycle angle $\phi = 2\pi$. The sense of rotation is reversed when the field reaches the x axis. The waveforms show the orthogonal components $H_{0,x}(t)$ (blue) and $H_{0,y}(t)$ (yellow), each with amplitude H_0 . Bottom: With finite relaxation, the magnetization lags behind the field, producing a cycle-averaged bias toward the y axis (perpendicular to the switching axis). Accordingly, the cycle-averaged pair potential favors dimer alignment along the y axis. (c) EARMF protocol and schematic illustration of the absence of orientational selectivity at the isotropic half-sweep angle $\phi = \phi_{iso}$. Top: $\mathbf{H}_0(t)$ sweeps a larger half-cycle angle $\phi = 2\pi + 2\alpha$ and reverses at the switching angles $\pm\alpha$ relative to the x axis. Bottom: Increasing the sweep angle makes the field sample directions near the switching axis more thoroughly, compensating for the missing contribution near x in the regular ARMF. A central goal of this work is to determine the isotropic half-sweep angle ϕ_{iso} at which the cycle-averaged pair potential becomes isotropic. At this angle, no dimer orientation is energetically preferred. (d) Definition of the dimer orientation angle $\theta \in [0, \pi)$ measured from the x axis. The dimer configurations shown in panels (b) and (c) are schematic illustrations and are not snapshots obtained from experiments or Brownian dynamics simulations.

model allows us to isolate the phase lag induced solely by finite relaxation. The additional effects of dipolar interaction fields and particle rotation are then examined numerically in Sec. III. In this limit, all particles have identical magnetization dynamics driven only by the external field, so we omit the particle index and write $\mathbf{m}_i(t) = \mathbf{m}(t)$. The simplified relaxation dynamics then obey [33,47]

$$\frac{d\mathbf{m}(t)}{dt} = -\frac{1}{\tau}(\mathbf{m}(t) - \chi_p \mathbf{H}_0(t)). \quad (4)$$

Solving this equation yields the following expressions for the steady-state Cartesian components of the magnetization (see Appendix A):

$$m_x(t) = \frac{\chi_p H_0}{1 + \tau^2 \omega^2} \left[\tau \omega \sin(\omega t - (2N_h + 1)\alpha) + \cos(\omega t - (2N_h + 1)\alpha) + \frac{2\tau \omega \sin \alpha}{1 - \exp\left(-\frac{2(\pi + \alpha)}{\tau \omega}\right)} \times \exp\left(-\frac{1}{\tau} \left[t - \frac{2(\pi + \alpha)N_h}{\omega} \right] \right) \right], \quad (5a)$$

$$m_y(t) = \frac{\chi_p H_0}{1 + \tau^2 \omega^2} (-1)^{N_h+1} \left[\tau \omega \cos(\omega t - (2N_h + 1)\alpha) - \sin(\omega t - (2N_h + 1)\alpha) - \frac{2\tau \omega \cos \alpha}{1 + \exp\left(-\frac{2(\pi + \alpha)}{\tau \omega}\right)} \times \exp\left(-\frac{1}{\tau} \left[t - \frac{2(\pi + \alpha)N_h}{\omega} \right] \right) \right]. \quad (5b)$$

Since $t' = t - N_h(T_{cyc}/2) = t - 2(\pi + \alpha)N_h/\omega$, the exponential factors in Eqs. (5a) and (5b) can be written as $\exp(-t'/\tau)$. Although these terms take an exponential form, they do not represent initial transients. Because the reduced time t' is reset at each half cycle, the same exponential variation is repeated in the periodic steady-state response.

Equation (5) shows that, for fixed τ , the magnetization amplitude decreases with increasing frequency. This frequency dependence also carries over to the overall scaling of the interaction energy derived below.

For two particles i and j , the centers of which are separated by r_{ij} with unit vector $\hat{\mathbf{r}}_{ij}$, the instantaneous dipolar interaction

is described as [48]

$$U_{ij}(t, \theta) = \frac{\mu_0}{4\pi r_{ij}^3} [\mathbf{m}_i(t) \cdot \mathbf{m}_j(t) - 3(\mathbf{m}_i(t) \cdot \hat{\mathbf{r}}_{ij})(\mathbf{m}_j(t) \cdot \hat{\mathbf{r}}_{ij})]. \quad (6)$$

Here, $\theta \in [0, \pi)$ denotes the angle between the dimer axis (the line of centers, $\hat{\mathbf{r}}_{ij}$) and the x axis [Fig. 1(d)]. When the field rotation is sufficiently fast that particles do not translate or reorient within one full period T_{cyc} , the cycle-averaged pair potential is

$$\langle U_{ij} \rangle(\theta) = \frac{1}{T_{\text{cyc}}} \int_0^{T_{\text{cyc}}} U_{ij}(t, \theta) dt. \quad (7)$$

Since the zero of $\langle U_{ij} \rangle(\theta)$ is arbitrary, we introduce the relative cycle-averaged pair potential referenced to $\theta = 0$ (alignment with the x axis),

$$\Delta \langle U_{ij} \rangle(\theta) = \langle U_{ij} \rangle(\theta) - \langle U_{ij} \rangle(0). \quad (8)$$

This definition removes the constant offset and leaves only the θ -dependent component that generates the aligning torque. We obtain (see Appendix B)

$$\begin{aligned} \Delta \langle U_{ij} \rangle(\theta) = & -\frac{3}{T_{\text{cyc}}} \frac{\mu_0}{4\pi r_{ij}^3} \left(\frac{\chi_p H_0}{1 + \tau^2 \omega^2} \right)^2 \frac{1}{\omega \sinh\left(\frac{\phi}{\tau \omega}\right)} \\ & \times \left[(1 + 3\tau^2 \omega^2) \sin(\phi) \sinh\left(\frac{\phi}{\tau \omega}\right) \right. \\ & \left. - 2\tau^3 \omega^3 \left\{ \cos(\phi) \cosh\left(\frac{\phi}{\tau \omega}\right) - 1 \right\} \right] (\cos(2\theta) - 1). \end{aligned} \quad (9)$$

Isotropy of the cycle-averaged interaction means that $\Delta \langle U_{ij} \rangle(\theta)$ is independent of the dimer orientation θ . Since $\Delta \langle U_{ij} \rangle(0) = 0$ by definition, isotropy is equivalent to $\Delta \langle U_{ij} \rangle(\theta) = 0$ for all θ . Therefore, the prefactor of $(\cos 2\theta - 1)$ in Eq. (9) must vanish. This condition yields the implicit equation defining the isotropic sweep angle ϕ_{iso} :

$$\begin{aligned} (1 + 3(\tau \omega)^2) \sin \phi_{\text{iso}} \sinh\left(\frac{\phi_{\text{iso}}}{\tau \omega}\right) \\ - 2(\tau \omega)^3 \left[\cos \phi_{\text{iso}} \cosh\left(\frac{\phi_{\text{iso}}}{\tau \omega}\right) - 1 \right] = 0. \end{aligned} \quad (10)$$

The regime $\tau \omega < \pi$ is of practical relevance for typical superparamagnetic colloids driven at experimentally accessible frequencies. In this regime, we derive a compact explicit approximation (see Appendix C)

$$\phi_{\text{iso,appr}} = 2\pi + \beta - \frac{4(\tau \omega)^3}{R} \exp\left(-\frac{2\pi + \beta}{\tau \omega}\right), \quad (11)$$

where

$$\begin{aligned} \beta &= \arctan\left(\frac{2(\tau \omega)^3}{1 + 3(\tau \omega)^2}\right), \\ R &= \sqrt{(1 + 3(\tau \omega)^2)^2 + (2(\tau \omega)^3)^2}. \end{aligned}$$

III. NUMERICAL CALCULATION

In Sec. II, we derived the isotropic sweep angle ϕ_{iso} under the assumption of single-timescale relaxation toward

$\chi_p \mathbf{H}_0(t)$. Here, we relax the noninteracting and nonrotating assumptions of the analytical model and numerically compute the magnetization dynamics, including the dipolar interaction field and particle rotation. Specifically, taking the analytical model used in Sec. II (with $\mathbf{H}_{L,i} = 0$ and $\boldsymbol{\Omega}_i = 0$) as a reference, we compare two numerical models: (1) a model including the interaction field $\mathbf{H}_{L,i}(t)$ but neglecting particle rotation ($\mathbf{H}_{L,i} \neq 0$, $\boldsymbol{\Omega}_i = 0$), and (2) a full model including both the interaction field and the rotational term ($\mathbf{H}_{L,i} \neq 0$, $\boldsymbol{\Omega}_i \neq 0$).

The time integration is performed using a forward Euler scheme applied to the nondimensionalized form of Eq. (2) (see Appendix D), with a uniform physical time step $\Delta t = 1.0 \times 10^{-6}$ s. The field angle is sampled on the same time grid. To ensure that the prescribed sweep angle is completed exactly within each half cycle, we set the number of steps per half cycle as $N_{\text{step},h} = \text{round}(\phi/(\omega \Delta t))$ and use a constant increment in the field angle $\Delta \phi_{\text{num}} = \phi/N_{\text{step},h}$. Since Δt is sufficiently small, this yields $\omega_{\text{num}} = \Delta \phi_{\text{num}}/\Delta t \approx \omega$. When particle rotation is included, the angular velocity $\boldsymbol{\Omega}_i(t)$ is determined from the balance between magnetic torque and rotational viscous drag in the overdamped limit. For a spherical particle of radius a in a fluid of viscosity η , the rotational drag coefficient is $\zeta_r = 8\pi \eta a^3$, and the angular velocity is given by $\boldsymbol{\Omega}_i(t) = \mathbf{T}_i(t)/\zeta_r$, where the magnetic torque is $\mathbf{T}_i(t) = \mu_0 \mathbf{m}_i(t) \times [\mathbf{H}_0(t) + \mathbf{H}_{L,i}(t)]$. In this study, the dimer configuration is kept fixed when evaluating the cycle-averaged pair potential. Therefore, the rotational term is not intended to describe the reorientation dynamics of the entire dimer. Rather, it is included to evaluate the effect of particle rotation on the magnetization dynamics and its impact on the isotropic condition.

The initial condition is set as $\mathbf{m}_i(0) = \mathbf{0}$. To ensure that transients have decayed regardless of the driving frequency, we discard the response over $0 \leq t < 5\tau$ and begin averaging at the start of the first complete cycle following $t = 5\tau$. After this transient regime, the response becomes periodic, and time averages are evaluated over a single subsequent period. We have confirmed that the steady-state results are independent of the initial condition and that averaging over multiple periods yields the same results within numerical precision.

For a given dimer orientation θ , the instantaneous interaction energy $U_{ij}(t, \theta)$ between the two induced dipoles is evaluated from Eq. (6), and the cycle-averaged pair potential is computed over one period as

$$\langle U_{ij} \rangle(\theta) = \frac{1}{T_{\text{cyc}}} \int_{t_0}^{t_0 + T_{\text{cyc}}} U_{ij}(t, \theta) dt, \quad (12)$$

where t_0 is taken in the steady-state regime. We then evaluate $\Delta \langle U_{ij} \rangle(\theta)$ as defined in Eq. (8).

Figure 2 shows the numerically obtained $\Delta \langle U_{ij} \rangle(\theta)$ for several half-sweep angles ϕ at $\tau = 4$ ms and $f = 40$ Hz ($\omega = 80\pi$ rad/s), computed using the model that includes the interaction field but neglects particle rotation. As ϕ increases beyond 2π , $\Delta \langle U_{ij} \rangle(\pi/2)$ changes from negative to positive at ϕ_{iso} , corresponding to a switch of the preferred dimer orientation from $\theta = \pi/2$ to $\theta = 0$. For these parameters, ϕ_{iso} lies between 380° and 390° . To numerically determine the isotropic half-sweep angle ϕ_{iso} for a given

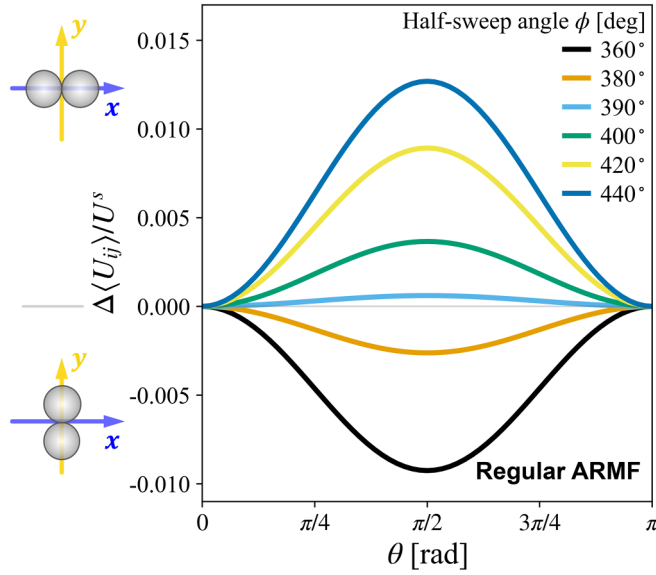


FIG. 2. Dimensionless relative potential $\Delta\langle U_{ij} \rangle(\theta)/U^s$ as a function of the dimer orientation angle θ for several half-sweep angles, computed using the numerical model including the interaction field but neglecting particle rotation ($\mathbf{H}_{I,i} \neq 0$, $\mathbf{\Omega}_i = 0$), for $\tau = 4$ ms and $f = 40$ Hz ($\omega = 80\pi$ rad/s). The energy normalization factor U^s is defined in Appendix D. If $\Delta\langle U_{ij} \rangle(\pi/2)$ is negative (positive), alignment along the y axis (x axis) is energetically favored. For these parameters, ϕ_{iso} lies between $\phi = 380^\circ$ and 390° .

pair (τ, ω) , we monitor $\Delta\langle U_{ij} \rangle(\pi/2)$ as a function of ϕ . Since $\Delta\langle U_{ij} \rangle(\pi/2) = \langle U_{ij} \rangle(\pi/2) - \langle U_{ij} \rangle(0)$, a sign change in $\Delta\langle U_{ij} \rangle(\pi/2)$ marks the point at which $\langle U_{ij} \rangle(\pi/2) = \langle U_{ij} \rangle(0)$. Over the scanned range $\phi > 2\pi$, $\Delta\langle U_{ij} \rangle(\pi/2)$ changes sign only once. Therefore, we locate ϕ_{iso} by linear interpolation between the two neighboring values of ϕ that straddle the sign change. Repeating this procedure while scanning ω yields $\phi_{\text{iso}}(\omega)$ for each relaxation time τ . The same qualitative trend, namely, a sign change of $\Delta\langle U_{ij} \rangle(\pi/2)$ as ϕ increases beyond 2π , is observed even when the rotational term is included. The resulting ϕ_{iso} values for the two models are systematically compared in Sec. IV.

IV. DISCUSSION

A. Isotropic sweep angle

In this subsection, we examine the master curve $\phi_{\text{iso}}(\tau\omega)$ obtained analytically and assess its robustness against the dipolar interaction field and particle rotation.

Figure 3(a) shows the isotropic half-sweep angle ϕ_{iso} as a function of the dimensionless parameter $\tau\omega$. As shown by Eq. (10), ϕ_{iso} depends on τ and ω only through the combination $\tau\omega$. Thus, the isotropic condition is represented by a master curve, $\phi_{\text{iso}}(\tau\omega)$. The solid line is the theoretical prediction from Eq. (10), the dotted line is the explicit approximation $\phi_{\text{iso,appr}}(\tau\omega)$ from Eq. (11), and the symbols are numerical results obtained for various (τ, ω) pairs using the model that includes the interaction field but neglects particle rotation.

In the analytical model, the magnetization relaxes only toward $\chi_p \mathbf{H}_0(t)$ [Eq. (4)]. In contrast, in the numerical model

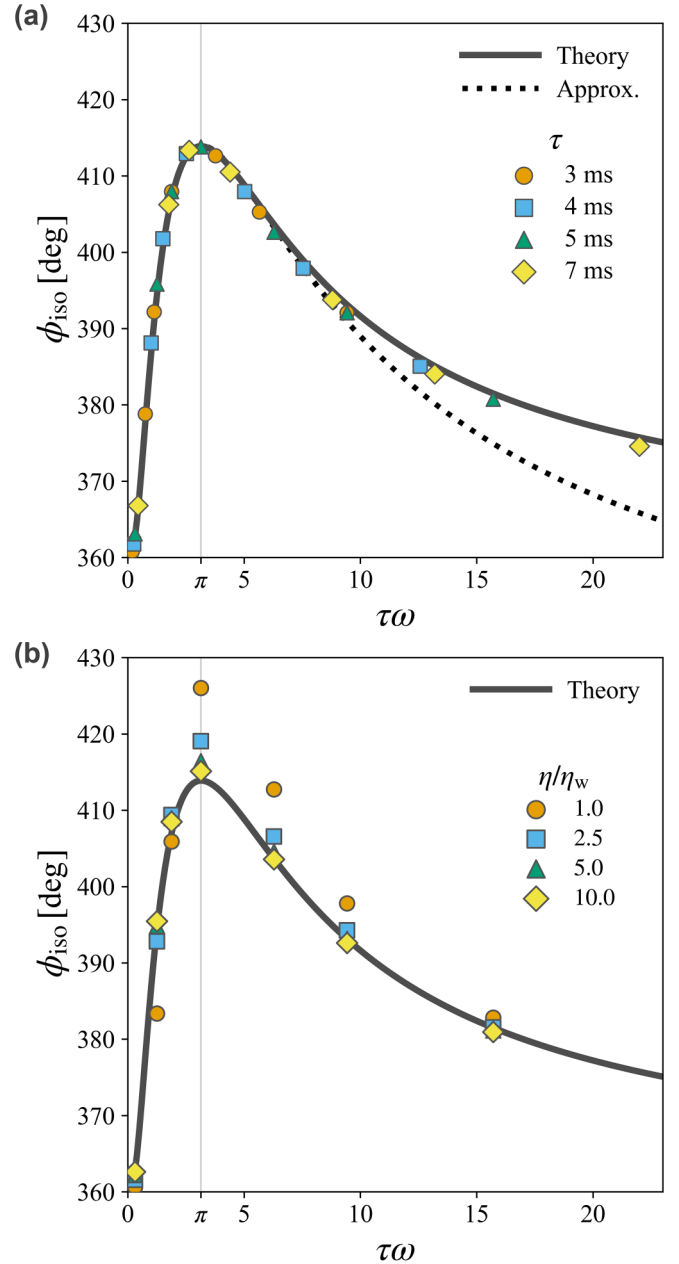


FIG. 3. (a) Isotropic sweep angle ϕ_{iso} as a function of $\tau\omega$. The implicit relation in Eq. (10) yields a master curve $\phi_{\text{iso}}(\tau\omega)$ with a peak near $\tau\omega \simeq \pi$ (solid line). Numerical results obtained by including only the interaction field while neglecting particle rotation (symbols) collapse onto this master curve with only minor deviations for various (τ, ω) pairs. The dotted line shows the explicit approximation in Eq. (11), which agrees well with the master curve for $\tau\omega < \pi$. (b) Comparison between the theoretical curve and numerical results including the rotational term. The relaxation time is fixed at $\tau = 5$ ms, and the viscosity ratio η/η_w is varied as 1, 2.5, 5, and 10, where $\eta_w = 0.896$ mPa s is the bulk water viscosity. As the viscosity increases, the contribution from particle rotation decreases, and the numerical results approach the theoretical curve.

including the interaction field but neglecting particle rotation, the magnetization relaxes toward $\chi_p(\mathbf{H}_0(t) + \mathbf{H}_{I,i}(t))$. Nevertheless, the numerically obtained ϕ_{iso} values lie close to the theoretical master curve, with only minor deviations. This

result indicates that, for a dimer, including the interaction field $\mathbf{H}_{I,i}(t)$ does not substantially modify the $\tau\omega$ dependence of ϕ_{iso} . Furthermore, in the experimentally relevant regime $\tau\omega < \pi$, the approximation reproduces the theoretical curve $\phi_{\text{iso}}(\tau\omega)$ with a relative error $|\phi_{\text{iso}} - \phi_{\text{iso,appr}}|/(\phi_{\text{iso}} - 2\pi)$ below 1%.

In the theoretical analysis in Sec. II and the numerical calculations in Sec. III, we held the dimer orientation fixed during each field cycle when evaluating the cycle-averaged pair potential. To test whether the orientational selectivity predicted using this fixed-orientation approximation is reproduced when the dimer is allowed to reorient dynamically, we performed overdamped Brownian dynamics simulations. In these simulations, the positions of the two constituent particles evolved at each time step according to the instantaneous forces and thermal fluctuations. The simulations were performed for initial orientations $\theta_0 = 0^\circ, 45^\circ$, and 90° , and for half-sweep angles below, near, and above ϕ_{iso} . The qualitative long-time orientational behavior was independent of the initial orientation. For $\phi < \phi_{\text{iso}}$, the dimer relaxed toward alignment along the y axis ($\theta = \pi/2$) and subsequently fluctuated around this orientation. For $\phi > \phi_{\text{iso}}$, the dimer relaxed toward alignment along the x axis ($\theta = 0$) and fluctuated around this orientation. In contrast, for $\phi \simeq \phi_{\text{iso}}$, no systematic orientational drift toward a particular direction was observed. Instead, the dimer orientation exhibited Brownian fluctuations without a preferred direction. The simulation model and numerical conditions are described in Appendix E, and the time evolution of the dimer orientation is shown in Fig. 6. The corresponding dimer dynamics are provided in the Supplemental Material for Movies [49].

Figure 3(b) shows the numerical results when the rotational term is also included. Because the effect of the rotational term depends on the viscosity of the medium, we examine its viscosity dependence at fixed $\tau = 5$ ms. The values of η range from 1 to 10 times the bulk water viscosity $\eta_w = 0.896$ mPa s.

For all viscosities, the qualitative shape of $\phi_{\text{iso}}(\tau\omega)$ and the position of its maximum are essentially preserved. Quantitatively, the deviation from the analytical curve becomes smaller as the viscosity increases, and the numerical results approach the analytical prediction. This behavior is consistent with the torque-balance relation $\mathbf{\Omega}_i = \mathbf{T}_i/\zeta_r$, which shows that the contribution of particle rotation decreases as the viscous resistance increases.

In typical experimental setups for paramagnetic colloids, the particles are denser than the surrounding solvent and are therefore expected to be located near the substrate. In this situation, hydrodynamic interactions with the substrate can reduce the rotational mobility associated with particle self-rotation compared with that of an isolated particle in a bulk fluid. In aggregated states, hydrodynamic interactions with neighboring particles may also suppress particle self-rotation. Therefore, the $\eta = \eta_w$ case in Fig. 3(b) provides a conservative upper bound on the effect of particle rotation on ϕ_{iso} .

The master curve $\phi_{\text{iso}}(\tau\omega)$ offers two practical advantages in experiments. First, for a chosen driving frequency, ϕ_{iso} serves as a convenient control parameter to toggle between isotropic and anisotropic effective interactions, providing a route to control colloidal morphologies and their dynamics under alternating fields. At $\phi = \phi_{\text{iso}}$, the half-cycle reversal

inherent to EARMF suppresses net cumulative rotation while maintaining an isotropic cycle-averaged pair interaction. This combination cannot be achieved by continuous-rotation protocols such as CRMF. Second, the master curve provides a general procedure for estimating an unknown relaxation time in other field-responsive systems whose induced-dipole response can be described by a single first-order relaxation time. At a fixed driving frequency ω , the half-sweep angle ϕ is varied while measuring an observable that quantifies the orientational anisotropy, such as the orientational order parameter $S_2 = \langle \cos 2\theta \rangle$. As ϕ is varied, the preferred orientation switches between two orthogonal directions through an isotropic point. The corresponding half-sweep angle, at which the orientational anisotropy vanishes and $S_2 = 0$, is identified as $\phi_{\text{iso}}(\omega)$. Repeating this procedure at several driving frequencies yields a set of $\phi_{\text{iso}}(\omega)$ values, which can then be fitted to the theoretical master curve $\phi_{\text{iso}}(\tau\omega)$ using τ as the fitting parameter. Measurements at multiple frequencies are important because the master curve is nonmonotonic, and a single value of ϕ_{iso} may therefore not uniquely determine τ .

B. Mechanism of the unimodal behavior of the isotropic sweep angle

As shown in Fig. 3, the extra sweep required for isotropy, $2\alpha_{\text{iso}} = \phi_{\text{iso}} - 2\pi$, has a single maximum near $\tau\omega = \pi$ rather than increasing monotonically with $\tau\omega$. To understand the origin of this peak, we analyze the cycle-averaged pair potential under a regular ARMF ($\phi = 2\pi$) and show that the anisotropy itself is unimodal in $\tau\omega$, accounting for the unimodal behavior of ϕ_{iso} . We further show that the peak of ϕ_{iso} does not coincide with the extremum of the anisotropy and explain this shift in terms of the monotonically decreasing magnetization amplitude with increasing $\tau\omega$.

We now derive an integral representation of the anisotropy that makes its $\tau\omega$ dependence explicit. The relaxation dynamics in Eq. (4) can also be written as a convolution of the instantaneous equilibrium magnetization $\mathbf{m}_{\text{eq}}(t) = \chi_p \mathbf{H}_0(t)$ with the relaxation kernel $k(u)$,

$$\begin{aligned} \mathbf{m}(t) &= \int_{-\infty}^{\infty} k(u) \mathbf{m}_{\text{eq}}(t-u) du, \\ k(u) &= \frac{1}{\tau} \exp\left(-\frac{u}{\tau}\right) \Theta(u), \end{aligned} \quad (13)$$

where $\Theta(u)$ is the Heaviside step function.

The anisotropic part of the cycle-averaged pair potential is defined as the difference between the potential at $\theta = \pi/2$ and at $\theta = 0$,

$$\Delta \langle U_{ij} \rangle_{\text{aniso}} = \Delta \langle U_{ij} \rangle \left(\frac{\pi}{2} \right) = \langle U_{ij} \rangle \left(\frac{\pi}{2} \right) - \langle U_{ij} \rangle(0), \quad (14)$$

and can be expressed as (see Appendix F)

$$\Delta \langle U_{ij} \rangle_{\text{aniso}} = -\frac{3\mu_0}{4\pi r_{ij}^3} \int_{-\infty}^{\infty} C_k(\Delta u) D_m(\Delta u) d\Delta u. \quad (15)$$

Here, $C_k(\Delta u)$ is the autocorrelation of the relaxation kernel $k(u)$,

$$C_k(\Delta u) = \int_{-\infty}^{\infty} k(v) k(v - \Delta u) dv = \frac{1}{2\tau} \exp\left(-\frac{|\Delta u|}{\tau}\right), \quad (16)$$

and $D_m(\Delta u)$ is determined by the driving waveform alone, through the autocorrelations of the equilibrium magnetization,

$$D_m(\Delta u) = C_{m,y}(\Delta u) - C_{m,x}(\Delta u),$$

$$C_{m,\sigma}(\Delta u) = \frac{1}{T_{\text{cyc}}} \int_0^{T_{\text{cyc}}} m_{\text{eq},\sigma}(t) m_{\text{eq},\sigma}(t - \Delta u) dt,$$

$$\sigma = x, y. \quad (17)$$

The time lag Δu appears as the common shift variable in both the kernel autocorrelation $C_k(\Delta u)$ and the equilibrium-magnetization autocorrelations $C_{m,\sigma}(\Delta u)$.

Although Eq. (15) is written in terms of the time lag Δu , it is convenient to introduce the phase lag $\Delta\xi = \omega\Delta u$, for which the $\tau\omega$ dependence becomes explicit. Using $\Delta\xi = \omega\Delta u$, $\xi = \omega t$, and $\omega T_{\text{cyc}} = 2\phi = 4\pi$ (under a regular ARMF), we can rewrite Eq. (15) as

$$\Delta\langle U_{ij} \rangle_{\text{aniso}} = -\frac{3\mu_0}{4\pi r_{ij}^3} \int_{-\infty}^{\infty} C_k(\tau\omega, \Delta\xi) D_m(\Delta\xi) d\Delta\xi, \quad (18)$$

where

$$C_k(\tau\omega, \Delta\xi) = \frac{1}{2\tau\omega} \exp\left(-\frac{|\Delta\xi|}{\tau\omega}\right),$$

$$D_m(\Delta\xi) = C_{m,y}(\Delta\xi) - C_{m,x}(\Delta\xi),$$

$$C_{m,\sigma}(\Delta\xi) = \frac{1}{4\pi} \int_0^{4\pi} m_{\text{eq},\sigma}(\xi) m_{\text{eq},\sigma}(\xi - \Delta\xi) d\xi,$$

$$\sigma = x, y.$$

Equation (18) makes clear that, for a regular ARMF waveform, the τ and ω dependence enters only through the combination $\tau\omega$. Since $C_k(\tau\omega, \Delta\xi)$ is an even function, and $D_m(\Delta\xi)$ is an even and 4π -periodic function, Eq. (18) can also be expressed as (see Appendix F)

$$\Delta\langle U_{ij} \rangle_{\text{aniso}} = -\frac{3\mu_0}{4\pi r_{ij}^3} \frac{1}{\tau\omega(1 - \exp(-\frac{4\pi}{\tau\omega}))} \times \int_0^{4\pi} \exp\left(-\frac{\Delta\xi}{\tau\omega}\right) D_m(\Delta\xi) d\Delta\xi. \quad (19)$$

The integral in Eq. (19) makes the origin of the unimodal behavior more transparent, as we now elaborate.

For a regular ARMF ($\phi = 2\pi$), a half-cycle time shift gives $m_{\text{eq},x}(t + T_{\text{cyc}}/2) = m_{\text{eq},x}(t)$, while $m_{\text{eq},y}(t + T_{\text{cyc}}/2) = -m_{\text{eq},y}(t)$. This half-cycle sign alternation in the y component makes the autocorrelation $C_{m,y}(\Delta\xi)$ vary more slowly with the phase lag $\Delta\xi$ than $C_{m,x}(\Delta\xi)$ [Fig. 4(a)]. Consequently, $D_m(\Delta\xi) = C_{m,y}(\Delta\xi) - C_{m,x}(\Delta\xi)$ changes sign from positive to negative and back to positive over one period $\Delta\xi \in [0, 4\pi)$. Moreover, $D_m(\Delta\xi)$ has zero average over one period, $\int_0^{4\pi} D_m(\Delta\xi) d\Delta\xi = 0$, which follows from the 4π periodicity and zero cycle averages of $m_{\text{eq},x}(\xi)$ and $m_{\text{eq},y}(\xi)$.

Figure 4(b) shows the weighted integrand $\exp(-\Delta\xi/\tau\omega) D_m(\Delta\xi)$ from Eq. (19) for several values of $\tau\omega$ (colored curves), together with $D_m(\Delta\xi)$ for reference (black curve) over one period ($0 \leq \Delta\xi < 4\pi$). This representation allows the anisotropy to be interpreted qualitatively in terms of the signed area under the weighted integrand curve for each $\tau\omega$. At small $\tau\omega$ (e.g., $\tau\omega = 0.1$), the exponential weight is sharply localized near $\Delta\xi = 0$, and the weighted integrand

is confined to a narrow short-lag region. The signed area is therefore small, with only a slight positive bias. At intermediate $\tau\omega$ (e.g., $\tau\omega = 2$), the weighted integrand broadens, allowing the first positive lobe of D_m to be sampled more fully. The negative lobe at larger $\Delta\xi$ is still suppressed by the exponential decay. The net positive area, and hence the magnitude of the (negative) anisotropy, is maximized in this regime. At large $\tau\omega$ (e.g., $\tau\omega = 10$), the exponential weight decays slowly, and the negative lobe contributes more substantially, partially canceling the positive short-lag contribution. In the limit $\tau\omega \rightarrow \infty$ ($\tau\omega = 100$ in practice), the weighting becomes nearly uniform, and the anisotropy vanishes since $\int_0^{4\pi} D_m d\Delta\xi = 0$.

This qualitative picture explains why $\Delta\langle U_{ij} \rangle_{\text{aniso}}$ has a minimum at an intermediate value of $\tau\omega$. Figure 4(c) shows $\Delta\langle U_{ij} \rangle_{\text{aniso}}/k_B T$ computed using $B_0 = 1.2$ mT (where $B_0 = \mu_0 H_0$), $a = 0.5$ μm , $r_{ij} = 2.1a$, $\chi = 0.96$, and $T = 298$ K, which set only the overall scale. Indeed, the anisotropy exhibits a clear minimum at $\tau\omega \simeq \pi/2$. Since a larger anisotropy in regular ARMF requires a larger compensating sweep, this also accounts for the unimodal behavior of ϕ_{iso} . A key observation here is that the value of $\tau\omega$ minimizing $\Delta\langle U_{ij} \rangle_{\text{aniso}}$ under regular ARMF ($\tau\omega \simeq \pi/2$) does not coincide with the value $\tau\omega \simeq \pi$ that maximizes ϕ_{iso} . This is because the isotropic sweep angle is determined not only by the magnitude of the anisotropic potential itself, but also by how effectively an increase in ϕ compensates for that anisotropy.

To understand this peak shift, we examine the contour plot of $\Delta\langle U_{ij} \rangle_{\text{aniso}}/k_B T$ on the $(\tau\omega, \phi)$ plane shown in Fig. 4(d). The same parameters as in Fig. 4(c) are used, and the cross section at $\phi = 2\pi$ corresponds to the results presented in Fig. 4(c). At small $\tau\omega$, the contours are closely spaced along the ϕ direction, indicating that a small change in ϕ produces a large change in the anisotropy. At large $\tau\omega$, the contours are more widely spaced, meaning that a larger sweep angle is required to achieve the same change in anisotropy. This widening reflects the monotonic decrease of $\langle |\mathbf{m}|^2 \rangle$ with $\tau\omega$ [inset of Fig. 4(c)]. As the relaxation lag suppresses the magnetization response, the same sweep increment $\Delta\phi$ generates a smaller change in the dipolar potential. Consequently, for two $\tau\omega$ values that yield the same $\Delta\langle U_{ij} \rangle_{\text{aniso}}$ at $\phi = 2\pi$, the additional sweep angle needed to cancel it is larger at the larger $\tau\omega$. This explains why the peak of ϕ_{iso} ($\tau\omega \simeq \pi$) is located at a larger $\tau\omega$ than the minimum of the anisotropy ($\tau\omega \simeq \pi/2$).

C. Validity of the fixed-orientation approximation

Throughout this work, we assumed that the dimer orientation changes negligibly within a half cycle. The validity of this assumption depends on experimental parameters such as the viscosity of the medium, the field strength, and the particle susceptibility. Here, we provide an order-of-magnitude estimate for the dimer rotation over a half cycle to clarify when this assumption is justified.

For a dimer driven by a CRMF in the asynchronous regime, $\omega > \omega_c$, the steady-state angular velocity is [33,50–53]

$$\omega_n = \omega - \sqrt{\omega^2 - \omega_c^2}, \quad (20)$$

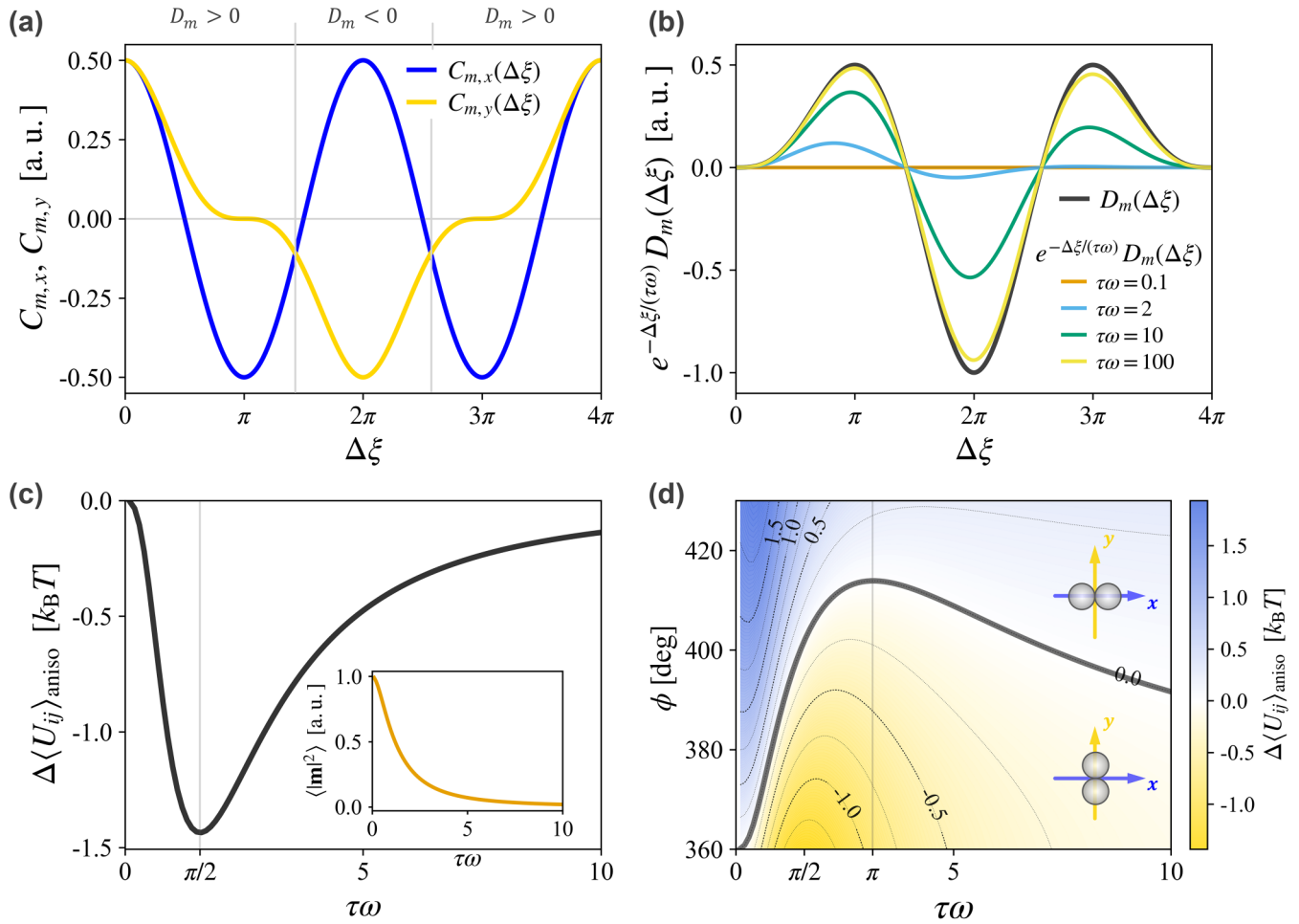


FIG. 4. (a) $C_{m,x}(\Delta\xi)$ and $C_{m,y}(\Delta\xi)$, the autocorrelations of the equilibrium-magnetization components $m_{\text{eq},x}$ and $m_{\text{eq},y}$ for a regular ARMF protocol ($\phi = 2\pi$), shown over the full cycle $0 \leq \Delta\xi < 4\pi$ (arbitrary units). (b) Colored curves show the integrand in Eq. (19), $e^{-\Delta\xi/(\tau\omega)} D_m(\Delta\xi)$, for $\tau\omega = 0.1, 2, 10, 100$ (arbitrary units). For reference, $D_m(\Delta\xi) = C_{m,y}(\Delta\xi) - C_{m,x}(\Delta\xi)$ is shown as the black curve. (c) Cycle-averaged anisotropic potential $\Delta\langle U_{ij} \rangle_{\text{aniso}}/k_B T$ as a function of $\tau\omega$ under regular ARMF, exhibiting a single minimum around $\tau\omega \simeq \pi/2$. The calculation uses $B_0 = 1.2$ mT, $a = 0.5$ μm , $r_{ij} = 2.1a$, $\chi = 0.96$, and $T = 298$ K. Inset shows the mean-squared magnetization $\langle |\mathbf{m}|^2 \rangle$ vs $\tau\omega$, which decreases monotonically as $\tau\omega$ increases due to the relaxation lag of $\mathbf{m}$ behind $\mathbf{H}_0$. (d) Contour plot of the cycle-averaged anisotropic potential $\Delta\langle U_{ij} \rangle_{\text{aniso}}/k_B T$ as a function of $\tau\omega$ and sweep angle ϕ , computed using the same parameters as in panel (c). The black line shows the isotropic sweep angle ϕ_{iso} , and the yellow region ($\Delta\langle U_{ij} \rangle_{\text{aniso}} < 0$) and blue region ($\Delta\langle U_{ij} \rangle_{\text{aniso}} > 0$) show areas where the dimer tends to align along the y and x axes, respectively.

where ω is the field angular frequency and ω_n is the dimer angular frequency. The critical angular frequency $\omega_c = 3\mu_0(\chi_p H_0)^2/(2\pi r_{ij}^5 \zeta)$ collects the relevant material properties and experimental parameters. Here, r_{ij} is the center-to-center distance of the dimer and $\zeta = 6\pi\eta a$ is the Stokes drag coefficient for a sphere of radius a in a fluid of viscosity η . In the limit $\omega \gg \omega_c$, a Taylor expansion gives

$$\frac{\omega_n}{\omega} \approx \frac{1}{2} \left(\frac{\omega_c}{\omega} \right)^2. \quad (21)$$

In CRMF, the field advances by 2π over one period, so the dimer rotates by an angle of order $2\pi(\omega_n/\omega)$ per period. In EARMF, in contrast, the field rotates in one direction only over a single half cycle before reversing in the next. As in the asynchronous CRMF regime, the field overtakes the slowly

rotating dimer, causing the magnetic torque to change sign and producing back-and-forth angular motion within a half cycle. Although a single EARMF half cycle does not correspond to the continuous steady-state rotation in CRMF, the underlying back-and-forth mechanism is similar. The CRMF steady-state angular velocity ω_n thus provides an order-of-magnitude estimate of the dimer rotation in EARMF. Since the field advances by ϕ during a half cycle, this estimate is of order $\phi(\omega_n/\omega)$, and the fixed-orientation approximation is expected to hold when $\phi(\omega_n/\omega) \ll 1$. For the parameters used in our numerical calculations ($B_0 = 1.2$ mT, $a = 0.5$ μm , $\chi = 0.96$, $r_{ij} = 2.1a$, $\eta = 0.896$ mPa s, and $f = 40$ Hz), we obtain $\omega_n/\omega \simeq 1.30 \times 10^{-3}$. For $\phi = 440^\circ$, which is the largest sweep angle per half cycle used in this work, this gives $\Delta\theta_{\text{half}} = \phi(\omega_n/\omega) \simeq 1.00 \times 10^{-2}$ rad, corresponding to 0.57° . Thus, the estimated rotation satisfies $\Delta\theta_{\text{half}} \ll 1$, supporting

the fixed-orientation approximation for the parameter range explored here.

Hydrodynamic interactions are not explicitly included in the fixed-orientation model used here. In general, translation-rotation coupling can quantitatively affect the rotational dynamics of a dimer. However, its effect under EARMF is expected to differ from that under CRMF. Under CRMF, the field continues to rotate in one direction, so even a small hydrodynamic contribution may accumulate over successive cycles. Under EARMF, in contrast, the field-rotation direction is reversed every half cycle. In the fast-driving regime considered here, the dimer configuration changes only slightly during one field cycle. Hydrodynamic effects associated with particle translation are therefore expected to produce only a small net change within a cycle. In addition, hydrodynamic motion caused by particle self-rotation should reverse when the field-rotation direction is reversed. If the particle configuration and hydrodynamic environment remain nearly unchanged, these contributions should largely cancel between the two half cycles. Hydrodynamic interactions may still affect the slower reorientation of the dimer over many cycles. They may also produce a quantitative shift in the experimentally measured value of ϕ_{iso} , particularly near confining walls or when lubrication effects become important. However, we do not expect them to alter the qualitative switching between the two orthogonal preferred orientations across the isotropic point.

V. CONCLUSION

Finite dipolar relaxation prevents the induced moment from tracking the applied field instantaneously. As a result, even a regular ARMF generally produces an anisotropic cycle-averaged pair potential.

In this work, we introduced the extended ARMF (EARMF), which extends the angular sweep beyond 2π , and analytically derived the additional sweep required to restore effective isotropy, both as an exact implicit relation and as a high-accuracy explicit approximation valid for $\tau\omega < \pi$. For a given relaxation time τ , the isotropy condition is frequency dependent and can be collapsed onto a master curve, $\phi_{\text{iso}} = \phi_{\text{iso}}(\tau\omega)$. Numerical simulations of the magnetization dynamics, including both the dipolar interaction term and particle rotation, confirm the analytical predictions. The implicit solution remains nearly exact when only the interaction term is included and reproduces the qualitative trends, including the unimodal behavior, when rotational dynamics are also accounted for. Importantly, $\phi_{\text{iso}}(\tau\omega)$ does not grow without bound as $\tau\omega$ increases. Instead, it reaches a maximum near $\tau\omega \simeq \pi$ and then decreases. This nontrivial behavior originates from the unimodal $\tau\omega$ dependence of the residual anisotropy under regular ARMF itself.

Although we used superparamagnetic colloids as a representative realization, the essential ingredients of the theory are only an induced dipole, a periodic field protocol, and a linear response with a single relaxation time for the dipolar dynamics. The same framework can therefore be adapted to other field-responsive soft matter systems with delayed polarization or magnetization, such as dielectric colloids driven by elec-

tric fields or ferrofluids with finite magnetic relaxation. More broadly, the analysis highlights how a phase-lagged response under time-periodic forcing can be used to tune effective anisotropy, a principle that may also be relevant to systems with frequency-dependent rotational or chiral responses.

Finally, the master curve for ϕ_{iso} offers practical advantages for experiments. For a chosen driving frequency, ϕ_{iso} can be used as a convenient control parameter to switch between isotropic and anisotropic cycle-averaged pair interactions. When τ is unknown, it can be estimated by measuring ϕ_{iso} at several driving frequencies and fitting the data to $\phi_{\text{iso}}(\tau\omega)$. In addition, at $\phi = \phi_{\text{iso}}$, the half-cycle reversal inherent in EARMF suppresses net rotation of both individual particles and clusters while maintaining an isotropic cycle-averaged pair interaction. This pair-level control may provide a useful starting point for future studies of slow many-particle processes, such as glass formation and crystal growth, for which assembly kinetics and many-body effects must also be considered.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

APPENDIX A: LAPLACE-TRANSFORM SOLUTION FOR THE DIPOLE DYNAMICS

For each Cartesian component of Eq. (4), the magnetization with finite relaxation time τ obeys

$$\frac{dm_{\sigma}(t)}{dt} = -\frac{1}{\tau}(m_{\sigma}(t) - \chi_p H_0 h_{\sigma}(t)), \quad \sigma = x, y. \quad (\text{A1})$$

The EARMF protocol [Eq. (1)] gives the driving functions $h_x(t)$ and $h_y(t)$ as

$$\begin{aligned} h_x(t) &= \cos(\omega t - (2N_h + 1)\alpha), \\ h_y(t) &= (-1)^{N_h} \sin(\omega t - (2N_h + 1)\alpha), \end{aligned} \quad (\text{A2})$$

where $N_h = \lfloor t/(T_{\text{cyc}}/2) \rfloor$ denotes the half-cycle index. One readily verifies that

$$h_x(t + T_{\text{cyc}}/2) = h_x(t), \quad h_y(t + T_{\text{cyc}}/2) = -h_y(t), \quad (\text{A3})$$

so $h_x(t)$ is periodic with period $T_{\text{cyc}}/2$, whereas $h_y(t)$ is antiperiodic with period $T_{\text{cyc}}/2$ (and hence periodic with period T_{cyc}). Here, $T_{\text{cyc}}/2 = \phi/\omega$, and ϕ is the field sweep angle over a half cycle.

The standard formula for the Laplace transform of a periodic function $p(t)$ with period T_{cyc} is

$$\mathcal{L}\{p(t)\}(s) = \frac{1}{1 - e^{-sT_{\text{cyc}}}} \int_0^{T_{\text{cyc}}} e^{-st} p(t) dt. \quad (\text{A4})$$

Substituting $h_x(t)$ and $h_y(t)$ into $p(t)$ and evaluating the integrals over one period $[0, T_{\text{cyc}})$, we obtain

$$\begin{aligned}\mathcal{L}\{h_x(t)\}(s) &= \frac{\omega \sin \alpha (1 + e^{-sT_{\text{cyc}}/2}) + s \cos \alpha (1 - e^{-sT_{\text{cyc}}/2})}{(\omega^2 + s^2)(1 - e^{-sT_{\text{cyc}}/2})}, \\ \mathcal{L}\{h_y(t)\}(s) &= \frac{\omega \cos \alpha (1 - e^{-sT_{\text{cyc}}/2}) - s \sin \alpha (1 + e^{-sT_{\text{cyc}}/2})}{(\omega^2 + s^2)(1 + e^{-sT_{\text{cyc}}/2})}.\end{aligned}\quad (\text{A5})$$

Taking the Laplace transform of Eq. (A1) and assuming $m_x(0) = m_y(0) = 0$, we obtain

$$\begin{aligned}\mathcal{L}\{m_x(t)\}(s) &= \frac{\chi_p H_0}{\tau} \frac{\omega \sin \alpha (1 + e^{-sT_{\text{cyc}}/2}) + s \cos \alpha (1 - e^{-sT_{\text{cyc}}/2})}{(s + 1/\tau)(\omega^2 + s^2)(1 - e^{-sT_{\text{cyc}}/2})}, \\ \mathcal{L}\{m_y(t)\}(s) &= \frac{\chi_p H_0}{\tau} \frac{\omega \cos \alpha (1 - e^{-sT_{\text{cyc}}/2}) - s \sin \alpha (1 + e^{-sT_{\text{cyc}}/2})}{(s + 1/\tau)(\omega^2 + s^2)(1 + e^{-sT_{\text{cyc}}/2})}.\end{aligned}\quad (\text{A6})$$

For Eq. (A6), the Laplace-domain expressions are decomposed into a purely rational part and a part multiplied by the factor $(1 \pm e^{-sT_{\text{cyc}}/2})/(1 \mp e^{-sT_{\text{cyc}}/2})$. The purely rational part is handled by partial-fraction decomposition and elementary inverse Laplace transforms. For the remaining part, we expand $(1 \mp e^{-sT_{\text{cyc}}/2})^{-1}$ as an infinite geometric series and then apply the time-shift theorem in the inverse Laplace transform. As a consequence, for a given time t , only a finite number of half-cycle contributions are nonzero, so the infinite series truncates to a finite sum.

After algebraic simplification, $m_x(t)$ and $m_y(t)$ are given by Eq. (A7) for $t \in [N_h T_{\text{cyc}}/2, (N_h + 1)T_{\text{cyc}}/2)$. The exponential terms retained in Eq. (A7) arise from the periodic resetting of the half-cycle boundary condition and are part of the steady-state periodic response. In contrast, the final terms in Eq. (A7) represent initial-condition transients, which decay on the timescale τ and become negligible when $t \gg \tau$:

$$\begin{aligned}m_x(t) &= \frac{\chi_p H_0}{1 + \tau^2 \omega^2} \left[\left\{ \tau \omega \sin(\omega t - (2N_h + 1)\alpha) + \cos(\omega t - (2N_h + 1)\alpha) + \frac{2\tau \omega \sin \alpha}{1 - \exp(-\frac{2(\pi + \alpha)}{\tau \omega})} \right. \right. \\ &\quad \times \exp\left(-\frac{1}{\tau} \left[t - \frac{2(\pi + \alpha)N_h}{\omega} \right] \right) \Big\} - \exp\left(-\frac{t}{\tau}\right) \left(\tau \omega \sin \alpha + \cos \alpha - \frac{2\tau \omega \sin \alpha}{1 - \exp(\frac{2(\pi + \alpha)}{\tau \omega})} \right) \Big], \\ m_y(t) &= \frac{\chi_p H_0}{1 + \tau^2 \omega^2} \left[(-1)^{N_h + 1} \left\{ \tau \omega \cos(\omega t - (2N_h + 1)\alpha) - \sin(\omega t - (2N_h + 1)\alpha) - \frac{2\tau \omega \cos \alpha}{1 + \exp(-\frac{2(\pi + \alpha)}{\tau \omega})} \right. \right. \\ &\quad \times \exp\left(-\frac{1}{\tau} \left[t - \frac{2(\pi + \alpha)N_h}{\omega} \right] \right) \Big\} - \exp\left(-\frac{t}{\tau}\right) \left(\tau \omega \cos \alpha - \sin \alpha - \frac{2\tau \omega \cos \alpha}{1 + \exp(\frac{2(\pi + \alpha)}{\tau \omega})} \right) \Big].\end{aligned}\quad (\text{A7})$$

These expressions will be useful to determine the time-averaged interaction potential between two dipoles.

APPENDIX B: CYCLE-AVERAGED PAIR POTENTIAL

The dipole-dipole interactions between particles i and j [Eq. (6)] can be written as

$$U_{ij}(t, \theta) = \frac{\mu_0}{4\pi r_{ij}^3} [|\mathbf{m}(t)|^2 - 3(\mathbf{m}(t) \cdot \hat{\mathbf{r}}_{ij})^2], \quad (\text{B1})$$

where we set $\mathbf{m}_i(t) = \mathbf{m}_j(t) = \mathbf{m}(t)$, because we neglect the dipolar field from neighboring particles in the magnetization dynamics [Eq. (4)]. Here, $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ and $\hat{\mathbf{r}}_{ij} = \mathbf{r}_{ij}/|\mathbf{r}_{ij}|$. We focus on the steady state and perform the cycle average over one period T_{cyc} using Eq. (7).

Within one cycle, the magnetization takes two forms,

$$\mathbf{m}(t) = \begin{cases} \mathbf{m}_0(t), & 0 \leq t < T_{\text{cyc}}/2, \\ \mathbf{m}_1(t), & T_{\text{cyc}}/2 \leq t < T_{\text{cyc}}, \end{cases} \quad (\text{B2})$$

so the cycle-averaged pair potential can be written as

$$\begin{aligned}\langle U_{ij} \rangle(\theta) &= \frac{1}{T_{\text{cyc}}} \frac{\mu_0}{4\pi r_{ij}^3} \\ &\times \left(\int_0^{T_{\text{cyc}}/2} (|\mathbf{m}_0(t)|^2 - 3(\mathbf{m}_0(t) \cdot \hat{\mathbf{r}}_{ij})^2) dt \right. \\ &\quad \left. + \int_{T_{\text{cyc}}/2}^{T_{\text{cyc}}} (|\mathbf{m}_1(t)|^2 - 3(\mathbf{m}_1(t) \cdot \hat{\mathbf{r}}_{ij})^2) dt \right). \quad (\text{B3})\end{aligned}$$

We parametrize the unit separation vector as $\hat{\mathbf{r}}_{ij} = (\cos \theta, \sin \theta)^\top$ and write the steady-state magnetizations as

$$\begin{aligned}\mathbf{m}_0(t) &= \frac{\chi_p H_0}{1 + \tau^2 \omega^2} \begin{pmatrix} \tilde{m}_{0,x}(t) \\ \tilde{m}_{0,y}(t) \end{pmatrix}, \\ \mathbf{m}_1(t) &= \frac{\chi_p H_0}{1 + \tau^2 \omega^2} \begin{pmatrix} \tilde{m}_{1,x}(t) \\ \tilde{m}_{1,y}(t) \end{pmatrix},\end{aligned}\quad (\text{B4})$$

where $\tilde{m}_{0,x}(t)$ and $\tilde{m}_{0,y}(t)$ are the dimensionless components obtained from Eq. (5) after factoring out $\chi_p H_0/(1 + \tau^2 \omega^2)$

and setting $N_h = 0$, while $\tilde{m}_{1,x}(t)$ and $\tilde{m}_{1,y}(t)$ are the dimensionless components obtained from Eq. (5) after factoring out $\chi_p H_0 / (1 + \tau^2 \Omega^2)$ and setting $N_h = 1$. By time-translation symmetry of the steady-state solution, the result is independent of the choice of consecutive half cycles.

Defining

$$\begin{aligned} f_0(t, \theta) &= \tilde{m}_{0,x}(t) \cos \theta + \tilde{m}_{0,y}(t) \sin \theta, \\ f_1(t, \theta) &= \tilde{m}_{1,x}(t) \cos \theta + \tilde{m}_{1,y}(t) \sin \theta, \\ |\tilde{\mathbf{m}}_0(t)|^2 &= \tilde{m}_{0,x}(t)^2 + \tilde{m}_{0,y}(t)^2, \\ |\tilde{\mathbf{m}}_1(t)|^2 &= \tilde{m}_{1,x}(t)^2 + \tilde{m}_{1,y}(t)^2, \end{aligned} \quad (\text{B5})$$

Eq. (B3) can be written as

$$\begin{aligned} \langle U_{ij} \rangle(\theta) &= \frac{1}{T_{\text{cyc}}} \frac{\mu_0}{4\pi r_{ij}^3} \left(\frac{\chi_p H_0}{1 + \tau^2 \omega^2} \right)^2 \\ &\times \left(\left[\int_0^{T_{\text{cyc}}/2} |\tilde{\mathbf{m}}_0(t)|^2 dt + \int_{T_{\text{cyc}}/2}^{T_{\text{cyc}}} |\tilde{\mathbf{m}}_1(t)|^2 dt \right] \right. \\ &\left. - 3 \left[\int_0^{T_{\text{cyc}}/2} f_0(t, \theta)^2 dt + \int_{T_{\text{cyc}}/2}^{T_{\text{cyc}}} f_1(t, \theta)^2 dt \right] \right). \end{aligned} \quad (\text{B6})$$

Here, we introduce the relative potential referenced to $\theta = 0$ (x axis direction),

$$\Delta \langle U_{ij} \rangle(\theta) = \langle U_{ij} \rangle(\theta) - \langle U_{ij} \rangle(0). \quad (\text{B7})$$

Since $|\tilde{\mathbf{m}}_0(t)|^2$ and $|\tilde{\mathbf{m}}_1(t)|^2$ do not depend on θ , we obtain

$$\begin{aligned} \Delta \langle U_{ij} \rangle(\theta) &= - \frac{3}{T_{\text{cyc}}} \frac{\mu_0}{4\pi r_{ij}^3} \left(\frac{\chi_p H_0}{1 + \tau^2 \omega^2} \right)^2 \\ &\times \left(\left[\int_0^{T_{\text{cyc}}/2} f_0(t, \theta)^2 dt + \int_{T_{\text{cyc}}/2}^{T_{\text{cyc}}} f_1(t, \theta)^2 dt \right] \right. \\ &\left. - \left[\int_0^{T_{\text{cyc}}/2} f_0(t, 0)^2 dt + \int_{T_{\text{cyc}}/2}^{T_{\text{cyc}}} f_1(t, 0)^2 dt \right] \right). \end{aligned} \quad (\text{B8})$$

Using the steady-state solution of $\mathbf{m}(t)$ [Eq. (5)], the integrals involving $f_0(t, \theta)$ and $f_1(t, \theta)$ can be evaluated as

$$\begin{aligned} &\int_0^{T_{\text{cyc}}/2} f_0(t, \theta)^2 dt + \int_{T_{\text{cyc}}/2}^{T_{\text{cyc}}} f_1(t, \theta)^2 dt \\ &= \left[\frac{(1 + \tau^2 \omega^2) \phi}{\omega} + \frac{2\tau^3 \omega^2}{\sinh\left(\frac{\phi}{\tau \omega}\right)} \left(\cosh\left(\frac{\phi}{\tau \omega}\right) - \cos \phi \right) \right] \\ &+ \frac{1}{\omega \sinh\left(\frac{\phi}{\tau \omega}\right)} \left[(1 + 3\tau^2 \omega^2) \sin \phi \sinh\left(\frac{\phi}{\tau \omega}\right) \right. \\ &\left. - 2\tau^3 \omega^3 \left(\cos \phi \cosh\left(\frac{\phi}{\tau \omega}\right) - 1 \right) \right] \cos(2\theta). \end{aligned} \quad (\text{B9})$$

Substituting Eq. (B9) into Eq. (B8), we obtain

$$\begin{aligned} \Delta \langle U_{ij} \rangle(\theta) &= - \frac{3}{T_{\text{cyc}}} \frac{\mu_0}{4\pi r_{ij}^3} \left(\frac{\chi_p H_0}{1 + \tau^2 \omega^2} \right)^2 \frac{1}{\omega \sinh\left(\frac{\phi}{\tau \omega}\right)} \\ &\times \left[(1 + 3\tau^2 \omega^2) \sin \phi \sinh\left(\frac{\phi}{\tau \omega}\right) \right. \\ &\left. - 2\tau^3 \omega^3 \left\{ \cos \phi \cosh\left(\frac{\phi}{\tau \omega}\right) - 1 \right\} \right] (\cos(2\theta) - 1), \end{aligned} \quad (\text{B10})$$

which corresponds to Eq. (9).

The cycle-averaged interaction is isotropic when $\Delta \langle U_{ij} \rangle(\theta)$ is independent of θ . Since $\Delta \langle U_{ij} \rangle(0) = 0$ by definition, isotropy is equivalent to

$$\Delta \langle U_{ij} \rangle(\theta) = 0, \quad \text{for all } \theta. \quad (\text{B11})$$

Therefore, the isotropy condition (B11) is equivalent to requiring the following coefficient to vanish:

$$\begin{aligned} &(1 + 3\tau^2 \omega^2) \sin \phi \sinh\left(\frac{\phi}{\tau \omega}\right) \\ &- 2\tau^3 \omega^3 \left(\cos \phi \cosh\left(\frac{\phi}{\tau \omega}\right) - 1 \right) = 0. \end{aligned}$$

This condition is identical to Eq. (10).

APPENDIX C: APPROXIMATE EXPLICIT EXPRESSION FOR THE ISOTROPIC SWEEP ANGLE

In this Appendix, we derive an explicit approximation for $\phi_{\text{iso}}(\tau \omega)$ that is accurate in the experimentally relevant regime $\tau \omega < \pi$. For convenience, we introduce the dimensionless variables in Eq. (10) as

$$x = \tau \omega > 0, \quad \phi_{\text{iso}} = 2\pi + 2\alpha_{\text{iso}}. \quad (\text{C1})$$

Using $\sin \phi_{\text{iso}} = \sin(2\alpha_{\text{iso}})$ and $\cos \phi_{\text{iso}} = \cos(2\alpha_{\text{iso}})$, and introducing $y = \phi_{\text{iso}}/x$, we obtain

$$(1 + 3x^2) \sinh(y) \sin(2\alpha_{\text{iso}}) - 2x^3 [\cosh(y) \cos(2\alpha_{\text{iso}}) - 1] = 0. \quad (\text{C2})$$

In this regime, y is large enough that we can apply $\sinh y \simeq \cosh y \simeq e^y/2$. Thus, Eq. (C2) reduces to

$$e^y [(1 + 3x^2) \sin(2\alpha_{\text{iso}}) - 2x^3 \cos(2\alpha_{\text{iso}})] + 4x^3 = 0. \quad (\text{C3})$$

Introducing

$$\begin{aligned} R &= [(1 + 3x^2)^2 + (2x^3)^2]^{1/2}, \\ \beta &= \arctan\left(\frac{2x^3}{1 + 3x^2}\right) \in (0, \pi/2), \end{aligned} \quad (\text{C4})$$

Eq. (C3) becomes

$$e^y \sin(2\alpha_{\text{iso}} - \beta) = -\frac{4x^3}{R}. \quad (\text{C5})$$

Defining a small deviation by

$$2\alpha_{\text{iso}} - \beta = \delta, \quad (\text{C6})$$

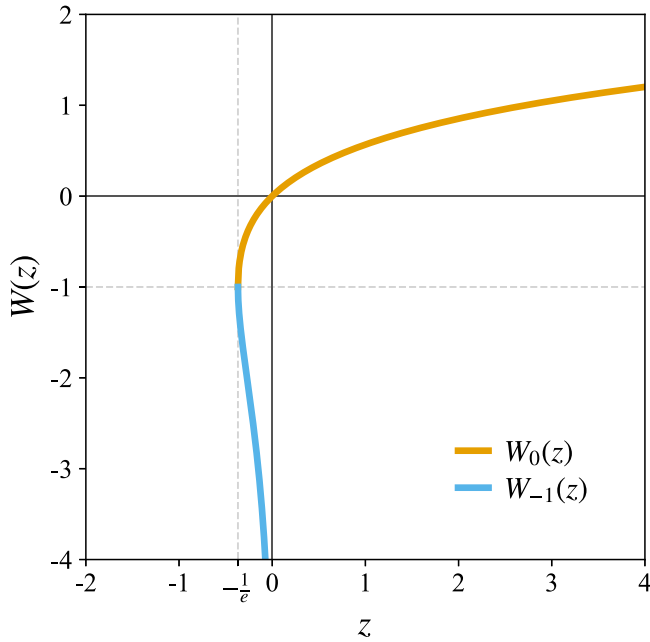


FIG. 5. Real branches of the Lambert W function on z : the principal branch $W_0(z)$ and the lower branch $W_{-1}(z)$, defined by $We^W = z$. At $z = -e^{-1}$, the two branches meet at $W = -1$.

we can write

$$y = \frac{\phi_{\text{iso}}}{x} = \frac{2\pi + \beta}{x} + \frac{\delta}{x}. \quad (\text{C7})$$

Substituting these into Eq. (C5) gives

$$\exp\left(\frac{\delta}{x}\right) \sin(\delta) = -\frac{4x^3}{R} \exp\left(-\frac{2\pi + \beta}{x}\right). \quad (\text{C8})$$

For small δ , we approximate $\sin \delta \simeq \delta$ and rearrange to

$$\left(\frac{\delta}{x}\right) \exp\left(\frac{\delta}{x}\right) = -\frac{4x^2}{R} \exp\left(-\frac{2\pi + \beta}{x}\right). \quad (\text{C9})$$

This can be solved by using the Lambert W function:

$$\frac{\delta}{x} = W\left(-\frac{4x^2}{R} \exp\left(-\frac{2\pi + \beta}{x}\right)\right), \quad (\text{C10})$$

where the Lambert W function is the multivalued inverse of $w \mapsto we^w$, defined by $W(z)e^{W(z)} = z$. For details on this function, see Sec. 4.13 of Ref. [54]. On the real axis, for $z \in [-e^{-1}, 0)$, two real branches exist: $W_0(z) \geq -1$ and $W_{-1}(z) < -1$ (Fig. 5).

In our solution,

$$\begin{aligned} \frac{\delta}{x} &= \frac{1}{x} \left(2\alpha_{\text{iso}} - \arctan\left(\frac{2x^3}{1+3x^2}\right) \right) \\ &> \frac{1}{x} \left(2\alpha_{\text{iso}} - \frac{2x^3}{1+3x^2} \right) \\ &= \left(\frac{2\alpha_{\text{iso}}}{x} + \frac{1+x^2}{1+3x^2} \right) - 1 \\ &> -1. \end{aligned} \quad (\text{C11})$$

Therefore, the physically admissible solution corresponds uniquely to the principal branch W_0 . Substituting back

$\delta = 2\alpha_{\text{iso}} - \beta$, we obtain

$$\begin{aligned} 2\alpha_{\text{iso}} &= \beta + x W_0\left(-\frac{4x^2}{R} \exp\left(-\frac{2\pi + \beta}{x}\right)\right), \\ \phi_{\text{iso}} &= 2\pi + 2\alpha_{\text{iso}} \\ &= 2\pi + \beta + x W_0\left(-\frac{4x^2}{R} \exp\left(-\frac{2\pi + \beta}{x}\right)\right). \end{aligned} \quad (\text{C12})$$

Since $W_0(z) = z - z^2 + O(z^3)$ near $z = 0$, and the argument is exponentially small for $x = \tau\omega < \pi$, we keep only the leading term,

$$\phi_{\text{iso,appr}} \simeq 2\pi + \beta - \frac{4x^3}{R} \exp\left(-\frac{2\pi + \beta}{x}\right). \quad (\text{C13})$$

Finally, substituting back $x = \tau\omega$, we recover the explicit elementary approximation given in Eq. (11).

APPENDIX D: NUMERICAL CALCULATION

For numerical stability and convenience, we integrate the magnetization dynamics in the nondimensional form of Eq. (2). This Appendix summarizes the nondimensionalization and the numerical procedure.

We choose the following characteristic scales: $H^s = H_0$ for magnetic-field amplitude, $a^s = a$ for length, $m^s = \chi_p H^s = 4\pi(a^s)^3 \chi H^s/3$ for magnetization, $t^s = 6\pi\eta(a^s)^3/(k_B T)$ for time, and $U^s = \mu_0 m^s H^s$ for energy. Here, H_0 is the field amplitude in A/m, a is the particle radius in m, χ is the (volume) magnetic susceptibility, η is the viscosity of the medium in Pa s, $k_B = 1.38 \times 10^{-23}$ J/K is the Boltzmann constant, T is the absolute temperature in K, and $\mu_0 = 4\pi \times 10^{-7}$ H/m is the vacuum permeability. Dimensionless variables are defined as $x^* = x/x^s$ (applied componentwise for vectors and tensors). In particular, $t^* = t/t^s$, $\tau^* = \tau/t^s$, and $\Delta t^* = \Delta t/t^s$.

Equation (2) can be written in nondimensional form as

$$\begin{aligned} \frac{d\mathbf{m}_i^*}{dt^*} &= -\frac{t^s}{\tau} (\mathbf{m}_i^* - (\mathbf{H}_0^* + \mathbf{H}_{l,i}^*)) \\ &\quad + \frac{\mu_0 m^s H^s t^s}{\zeta_r} (\mathbf{m}_i^* \times (\mathbf{H}_0^* + \mathbf{H}_{l,i}^*)) \times \mathbf{m}_i^*. \end{aligned} \quad (\text{D1})$$

Here, $\mathbf{H}_{l,i}^*$ is the dimensionless field induced by neighboring particles evaluated at the position of particle i , given in our nondimensionalization by

$$\mathbf{H}_{l,i}^* = \frac{\chi}{3|\mathbf{r}_{ij}^*|^3} [3\hat{\mathbf{r}}_{ij}\hat{\mathbf{r}}_{ij} - \mathbf{I}] \cdot \mathbf{m}_j^*, \quad (\text{D2})$$

where $\mathbf{r}_{ij}^* = \mathbf{r}_i^* - \mathbf{r}_j^*$, $\hat{\mathbf{r}}_{ij} = \mathbf{r}_{ij}^*/|\mathbf{r}_{ij}^*|$ and $\mathbf{I}$ is the identity matrix. Also, $\zeta_r = 8\pi\eta a^3$ denotes the rotational drag coefficient.

These equations are used to compute the time evolution of $\mathbf{m}_i^*$ with the initial condition $\mathbf{m}_i^*(0) = \mathbf{0}$. We integrate the dimensionless equation using a forward Euler scheme,

$$\mathbf{m}_i^*(t^* + \Delta t^*) = \mathbf{m}_i^*(t^*) + \Delta t^* \frac{d\mathbf{m}_i^*}{dt^*}. \quad (\text{D3})$$

Thus, the nondimensional discretized form of Eq. (2) becomes

$$\begin{aligned} \mathbf{m}_i^*(t^* + \Delta t^*) &= \mathbf{m}_i^*(t^*) - c_\tau^* [\mathbf{m}_i^*(t^*) - (\mathbf{H}_0^*(t^*) + \mathbf{H}_{l,i}^*(t^*))] \\ &\quad + c_r^* [(\mathbf{m}_i^*(t^*) \times (\mathbf{H}_0^*(t^*) \\ &\quad + \mathbf{H}_{l,i}^*(t^*))) \times \mathbf{m}_i^*(t^*)], \end{aligned} \quad (\text{D4})$$

where $c_\tau^* = \Delta t^* t^s / \tau = \Delta t / \tau$ and $c_r^* = \mu_0 m^s H^s t^s \Delta t^* / \zeta_r = \mu_0 m^s H^s \Delta t / \zeta_r$. The dipolar interaction potential in Eq. (6) is written in nondimensional form as

$$U_{ij}^*(t^*, \theta) = \frac{\chi}{3|\mathbf{r}_{ij}^*|^3} (\mathbf{m}_i^*(t^*) \cdot \mathbf{m}_j^*(t^*) - 3(\mathbf{m}_i^*(t^*) \cdot \hat{\mathbf{r}}_{ij})(\mathbf{m}_j^*(t^*) \cdot \hat{\mathbf{r}}_{ij})). \quad (\text{D5})$$

To compute the cycle-averaged potential in the steady state, we discard the response over $0 \leq t < 5\tau$ as a frequency-independent transient window, and then average $U_{ij}^*(t^*, \theta)$ over the first complete cycle starting after $t = 5\tau$:

$$\langle U_{ij}^* \rangle(\theta) = \frac{1}{T_{\text{cyc}}^*} \int_{t_0^*}^{t_0^* + T_{\text{cyc}}^*} U_{ij}^*(t^*, \theta) dt^*, \quad (\text{D6})$$

where $T_{\text{cyc}}^* = T_{\text{cyc}}/t^s$ and t_0^* denotes the start time of the averaging window. As stated in the main text, we assume that the particle configuration does not translate or reorient within one cycle.

We define the dimensionless relative cycle-averaged potential as $\Delta \langle U_{ij}^* \rangle(\theta) = \langle U_{ij}^* \rangle(\theta) - \langle U_{ij}^* \rangle(0)$, using the angle θ between the x axis and the line connecting the two particles. The numerical results shown in Figs. 2 and 3 were obtained as follows. We numerically determined ϕ_{iso} for all combinations of $\tau = 3, 4, 5, 7$ ms and $f = \omega/2\pi = 10, 40, 60, 100, 200, 300, 500$ Hz. For each pair of (τ, ω) , we computed $\Delta \langle U_{ij}^* \rangle(\theta)$ for half-sweep angles ϕ from 360° to 440° in steps of 10° . We located ϕ_{iso} by linear interpolation between the two neighboring values of ϕ that bracket the sign change of $\Delta \langle U_{ij}^* \rangle(\pi/2)$. In Fig. 3(a), we plotted ϕ_{iso} determined for all (τ, ω) pairs against $\tau\omega$. In these simulations, we used the following physical parameters before nondimensionalization: $T = 298$ K, $H_0 = 955$ A/m ($B_0 = \mu_0 H_0 = 1.2$ mT), $a = 0.5$ μm , $\chi = 0.96$, and $\eta = 0.896$ mPa s. In Fig. 3(b), we used the bulk water viscosity $\eta_w = 0.896$ mPa s and three larger values, $2.5\eta_w$, $5\eta_w$, and $10\eta_w$. In all numerical calculations above, as stated in the main text, the center-to-center distance was fixed at $2.1a$, and the time step was $\Delta t = 1.0 \times 10^{-6}$ s.

APPENDIX E: BROWNIAN DYNAMICS OF A DIMER

In this Appendix, we simulated the time-resolved dynamics of two interacting superparamagnetic particles forming a dimer using overdamped Brownian dynamics. The purpose of these simulations was to test whether the orientational selectivity predicted under the fixed-orientation approximation adopted in Secs. II and III could be qualitatively reproduced when the translational motion of the constituent particles was explicitly tracked. The particles were assumed to be suspended in a Newtonian fluid and to experience isotropic Stokes drag. At the micrometer scale considered here, particle inertia was negligible relative to viscous drag and was therefore neglected. The particle centers were constrained to move in the xy plane. Hydrodynamic interactions between the particles and hydrodynamic coupling to the confining wall were also neglected. To isolate dimer reorientation arising from the translational motion of its constituent particles, we omitted the rotational term $\boldsymbol{\Omega}_i \times \mathbf{m}_i$, which accounts for the rotation of the magnetic moment due to the rigid-body

self-rotation of particle i . Thus, individual particle self-rotation was neglected, whereas the dimer axis was free to reorient through the translational motion of the two particles. At each time step, the induced magnetic moment of particle i was updated using an explicit Euler scheme to describe relaxation toward $\chi_p[\mathbf{H}_0(t) + \mathbf{H}_{L,i}(t)]$ with relaxation time τ , while setting $\boldsymbol{\Omega}_i = 0$. The particle positions were then updated according to the overdamped Langevin equation using the Euler-Maruyama scheme,

$$\mathbf{r}_i^{n+1} = \mathbf{r}_i^n + \frac{\Delta t}{\zeta} \mathbf{F}_i^{\text{det},n} + \sqrt{\frac{2k_B T \Delta t}{\zeta}} \boldsymbol{\xi}_i^n, \quad (\text{E1})$$

where $\zeta = 6\pi\eta a$ is the translational Stokes drag coefficient. The vector $\boldsymbol{\xi}_i^n$ consists of independent standard normal random variables in the x and y directions and satisfies

$$\langle \xi_{i,\alpha}^n \rangle = 0, \quad \langle \xi_{i,\alpha}^n \xi_{j,\beta}^m \rangle = \delta_{ij} \delta_{\alpha\beta} \delta_{nm}, \quad (\text{E2})$$

where $\alpha, \beta \in \{x, y\}$, and the angle brackets denote an ensemble average over independent random realizations. The deterministic force acting on particle i was taken as the sum of the magnetic dipole-dipole force $\mathbf{F}_i^{\text{mag}}$, the short-range Hertzian contact force $\mathbf{F}_i^{\text{Hertz}}$, and the electrostatic repulsive force $\mathbf{F}_i^{\text{ele}}$,

$$\mathbf{F}_i^{\text{det}} = \mathbf{F}_i^{\text{mag}} + \mathbf{F}_i^{\text{Hertz}} + \mathbf{F}_i^{\text{ele}}. \quad (\text{E3})$$

The magnetic dipole-dipole force was evaluated from the instantaneous magnetic moments as

$$\begin{aligned} \mathbf{F}_i^{\text{mag}} = \frac{3\mu_0}{4\pi r_{ij}^4} [(\mathbf{m}_i \cdot \hat{\mathbf{r}}_{ij})\mathbf{m}_j + (\mathbf{m}_j \cdot \hat{\mathbf{r}}_{ij})\mathbf{m}_i \\ + (\mathbf{m}_i \cdot \mathbf{m}_j)\hat{\mathbf{r}}_{ij} - 5(\mathbf{m}_i \cdot \hat{\mathbf{r}}_{ij})(\mathbf{m}_j \cdot \hat{\mathbf{r}}_{ij})\hat{\mathbf{r}}_{ij}]. \end{aligned} \quad (\text{E4})$$

The short-range Hertzian contact force used to prevent particle overlap was written as

$$\mathbf{F}_i^{\text{Hertz}} = \begin{cases} \frac{4}{3} \frac{E}{2(1-\nu^2)} \left(\frac{a}{2}\right)^{\frac{1}{2}} (2a - r_{ij})^{\frac{3}{2}} \hat{\mathbf{r}}_{ij}, & r_{ij} < 2a, \\ 0, & r_{ij} \geq 2a, \end{cases} \quad (\text{E5})$$

where the Young's modulus and Poisson's ratio were set to $E = 1.0$ GPa and $\nu = 0.3$, respectively. The electrostatic repulsive force between the charged particle surfaces was calculated as

$$\mathbf{F}_i^{\text{ele}} = \frac{64\pi k_B T a \rho_\infty \gamma^2}{\kappa} \exp[-\kappa(r_{ij} - 2a)] \hat{\mathbf{r}}_{ij}, \quad (\text{E6})$$

where ρ_∞ is the bulk number density of ions, κ is the inverse Debye length, and $\gamma = \tanh(ze\psi_0/4k_B T)$. Here, z is the ionic valence, e is the elementary charge, and ψ_0 is the surface potential. The electrolyte was taken to be a 10 mM monovalent NaCl solution, such that $z = 1$ and $\kappa^{-1}(\text{nm}) = 0.304/\sqrt{c_{\text{salt}}}$, where c_{salt} is the concentration in M. The surface potential was set to $\psi_0 = -50$ mV. The simulations were performed using the following parameters. The particle radius was set to $a = 1.0$ μm , the magnetic susceptibility was $\chi = 0.96$, and the magnetic relaxation time was $\tau = 4.0$ ms. Within the present model, the particle radius does not enter Eq. (10) and therefore does not alter the theoretical isotropic half-sweep angle, although it affects the magnetic interaction strength, viscous drag, and timescale of the Brownian dynamics. The fluid viscosity was $\eta = 0.896$ mPa s. The driving frequency and

TABLE I. Parameters for Brownian dynamics simulations.

Category	Parameter	Value
Experimental conditions	ϕ (deg)	360, 386.7, 410
	T (K)	298
	f (Hz)	40
	B_0 (mT)	1.2
Superparamagnetic particles	a (μm)	1.0
	χ (—)	0.96
	τ (ms)	4.0
Hard sphere forces	E (GPa)	1.0
	ν (—)	0.3
Electrostatic forces	c_{salt} (mM)	10
	z (—)	1
	ψ_0 (mV)	−50.0
Medium properties	η (mPa s)	0.896

magnetic flux-density amplitude were fixed at $f = 40$ Hz and $B_0 = \mu_0 H_0 = 1.2$ mT, respectively, and the absolute

temperature was $T = 298$ K. Table I summarizes the simulation parameters.

The integration time step and total simulation duration were $\Delta t = 1.0 \times 10^{-6}$ s and $t_{\text{max}} = 50$ s, respectively. The initial center-to-center separation was $r_{ij}(t = 0) = 2.1a$. The governing equations were nondimensionalized before numerical integration. For these parameters, Eq. (10) predicts a theoretical isotropic half-sweep angle of $\phi_{\text{iso}}^{\text{th}} \simeq 386.7^\circ$. We therefore examined three half-sweep angles: $\phi = 360^\circ < \phi_{\text{iso}}^{\text{th}}$, $\phi = 386.7^\circ \simeq \phi_{\text{iso}}^{\text{th}}$, and $\phi = 410^\circ > \phi_{\text{iso}}^{\text{th}}$. For each half-sweep angle, the initial dimer orientation was set to $\theta_0 = 0^\circ$, 45° , or 90° . Figure 6 shows the time evolution of the simulated dimer orientation angle. For $\phi = 360^\circ < \phi_{\text{iso}}^{\text{th}}$, the dimer relaxed toward $\theta = \pi/2$, regardless of its initial orientation, and subsequently fluctuated around this preferred orientation. For $\phi = 410^\circ > \phi_{\text{iso}}^{\text{th}}$, the dimer instead relaxed toward $\theta = 0$ and fluctuated around this orientation. At $\phi = 386.7^\circ \simeq \phi_{\text{iso}}^{\text{th}}$, no systematic orientational drift toward a particular direction was observed. Instead, the dimer orientation exhibited Brownian fluctuations without selecting a preferred orientation. The cor-

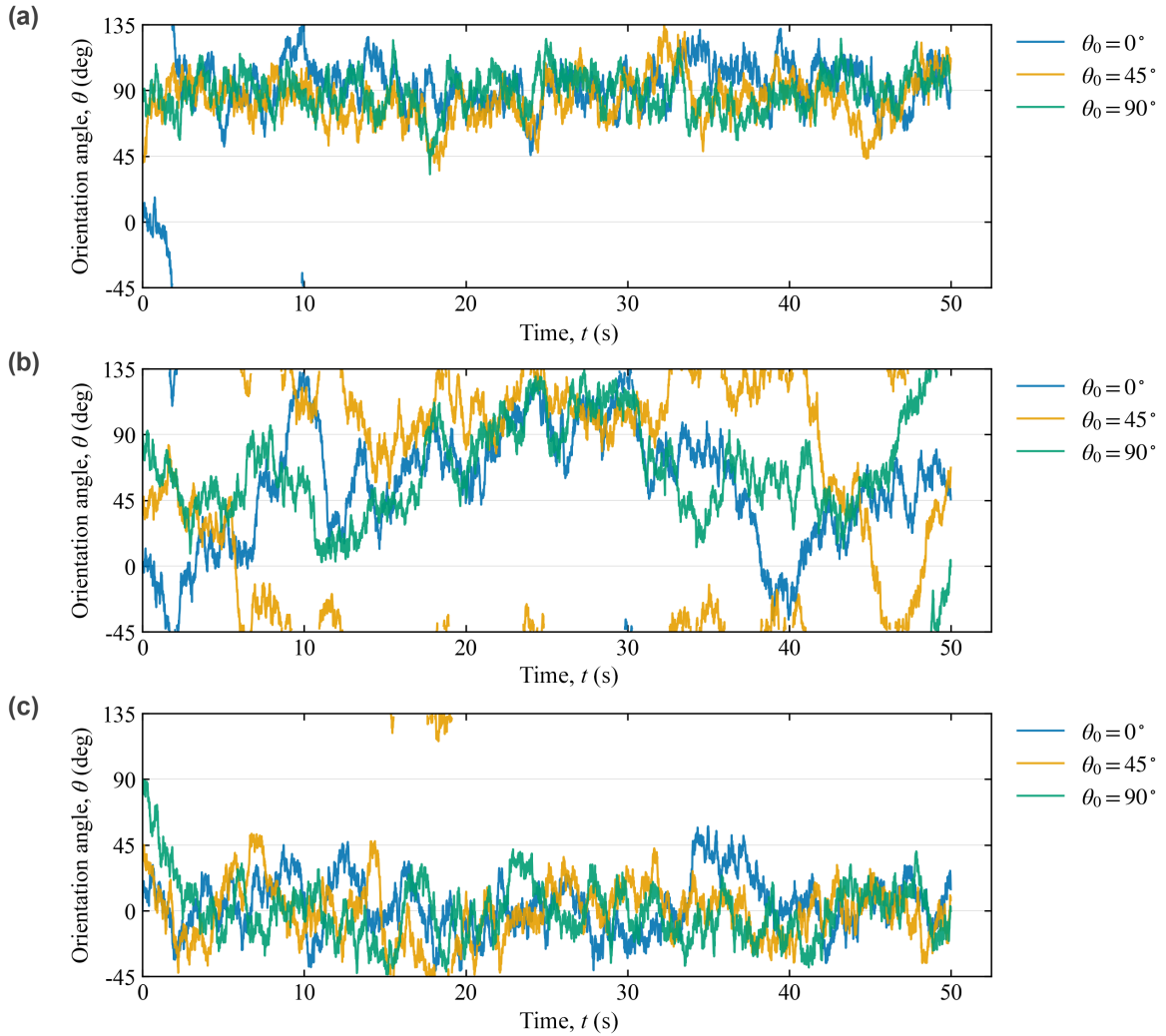


FIG. 6. Time evolution of the simulated dimer orientation angle θ for (a) $\phi = 360^\circ < \phi_{\text{iso}}^{\text{th}}$, (b) $\phi = 386.7^\circ \simeq \phi_{\text{iso}}^{\text{th}}$, and (c) $\phi = 410^\circ > \phi_{\text{iso}}^{\text{th}}$. For each half-sweep angle, simulations were initialized at $\theta_0 = 0^\circ$, 45° , and 90° , with an initial center-to-center separation of $r_{ij} = 2.1a$. For visualization, θ was wrapped to the interval $[-45^\circ, 135^\circ]$, and orientations differing by 180° represent the same dimer axis.

responding time-resolved dimer dynamics are provided in the Supplemental Material for Movies [49].

APPENDIX F: CONVOLUTION REPRESENTATION OF ANISOTROPIC POTENTIAL

In this Appendix, we derive the anisotropic component of the cycle-averaged pair potential, $\Delta\langle U_{ij} \rangle_{\text{aniso}} = \langle U_{ij} \rangle(\pi/2) - \langle U_{ij} \rangle(0)$, under a regular ARMF ($\phi = 2\pi$, so that $\omega T_{\text{cyc}} = 4\pi$). First, the magnetic dipole-dipole interaction between two identical moments $\mathbf{m}_i(t) = \mathbf{m}_j(t) = \mathbf{m}(t)$ separated by a distance r_{ij} reads

$$U_{ij}(t, \theta) = \frac{\mu_0}{4\pi r_{ij}^3} (|\mathbf{m}(t)|^2 - 3(\mathbf{m}(t) \cdot \hat{\mathbf{r}}_{ij})^2). \quad (\text{F1})$$

Writing $\mathbf{m}(t) = (m_x(t), m_y(t))^T$ and $\hat{\mathbf{r}}_{ij} = (\cos \theta, \sin \theta)^T$ gives

$$\begin{aligned} |\mathbf{m}(t)|^2 &= m_x(t)^2 + m_y(t)^2, \\ (\mathbf{m}(t) \cdot \hat{\mathbf{r}}_{ij})^2 &= m_x(t)^2 \cos^2 \theta + 2m_x(t)m_y(t) \sin \theta \cos \theta \\ &\quad + m_y(t)^2 \sin^2 \theta. \end{aligned} \quad (\text{F2})$$

We define the cycle-averaged pair potential $\langle U_{ij} \rangle(\theta)$ over one cycle T_{cyc} as

$$\langle U_{ij} \rangle(\theta) = \frac{1}{T_{\text{cyc}}} \int_0^{T_{\text{cyc}}} U_{ij}(t, \theta) dt. \quad (\text{F3})$$

Since $m_x(t)$ is periodic with period $T_{\text{cyc}}/2$ and $m_y(t)$ is antiperiodic with period $T_{\text{cyc}}/2$ [Eq. (5)], we have $\langle m_x(t)m_y(t) \rangle = 0$. Hence,

$$\begin{aligned} \langle |\mathbf{m}(t)|^2 \rangle &= \langle m_x(t)^2 \rangle + \langle m_y(t)^2 \rangle, \\ \langle (\mathbf{m}(t) \cdot \hat{\mathbf{r}}_{ij})^2 \rangle &= \langle m_x(t)^2 \rangle \cos^2 \theta + \langle m_y(t)^2 \rangle \sin^2 \theta. \end{aligned} \quad (\text{F4})$$

Therefore, substituting and rearranging gives

$$\begin{aligned} \langle U_{ij} \rangle &= -\frac{\mu_0}{4\pi r_{ij}^3} \left(\frac{1}{2} (\langle m_x(t)^2 \rangle + \langle m_y(t)^2 \rangle) \right. \\ &\quad \left. + \frac{3}{2} (\langle m_x(t)^2 \rangle - \langle m_y(t)^2 \rangle) \cos 2\theta \right). \end{aligned} \quad (\text{F5})$$

Using the definition of $\Delta\langle U_{ij} \rangle_{\text{aniso}}$, we obtain

$$\Delta\langle U_{ij} \rangle_{\text{aniso}} = -\frac{3\mu_0}{4\pi r_{ij}^3} (\langle m_y(t)^2 \rangle - \langle m_x(t)^2 \rangle). \quad (\text{F6})$$

Each component of Eq. (4) can also be written as a convolution of the equilibrium magnetization $\mathbf{m}_{\text{eq}}(t) = \chi_p \mathbf{H}_0(t)$ with the relaxation kernel

$$k(u) = \frac{1}{\tau} \exp\left(-\frac{u}{\tau}\right) \Theta(u), \quad (\text{F7})$$

where $\Theta(u)$ is the Heaviside step function. Thus,

$$m_\sigma(t) = \int_{-\infty}^{\infty} k(u) m_{\text{eq},\sigma}(t-u) du, \quad \sigma = x, y, \quad (\text{F8})$$

which corresponds to Eq. (13). The cycle-averaged squared magnetization $\langle m_\sigma(t)^2 \rangle$ can be expressed as

$$\begin{aligned} \langle m_\sigma(t)^2 \rangle &= \int_{-\infty}^{\infty} du_1 \int_{-\infty}^{\infty} du_2 \left[k(u_1)k(u_2) \frac{1}{T_{\text{cyc}}} \right. \\ &\quad \left. \times \int_0^{T_{\text{cyc}}} dt m_{\text{eq},\sigma}(t-u_1) m_{\text{eq},\sigma}(t-u_2) \right]. \end{aligned} \quad (\text{F9})$$

Introducing $\Delta u = u_1 - u_2$ and $v = (u_1 + u_2)/2$ and using the T_{cyc} periodicity of $m_{\text{eq},\sigma}(t)$, we obtain

$$\begin{aligned} \langle m_\sigma(t)^2 \rangle &= \int_{-\infty}^{\infty} d\Delta u \left[C_{m,\sigma}(\Delta u) \right. \\ &\quad \left. \times \int_{-\infty}^{\infty} dv k\left(v + \frac{\Delta u}{2}\right) k\left(v - \frac{\Delta u}{2}\right) \right], \end{aligned} \quad (\text{F10})$$

where

$$\begin{aligned} C_{m,\sigma}(\Delta u) &= \frac{1}{T_{\text{cyc}}} \int_0^{T_{\text{cyc}}} m_{\text{eq},\sigma}(t) m_{\text{eq},\sigma}(t - \Delta u) dt, \\ \sigma &= x, y. \end{aligned} \quad (\text{F11})$$

Defining the kernel autocorrelation

$$C_k(\Delta u) = \int_{-\infty}^{\infty} dv k(v) k(v - \Delta u) = \frac{1}{2\tau} \exp\left(-\frac{|\Delta u|}{\tau}\right), \quad (\text{F12})$$

we obtain the compact convolution form

$$\langle m_\sigma(t)^2 \rangle = \int_{-\infty}^{\infty} C_k(\Delta u) C_{m,\sigma}(\Delta u) d\Delta u. \quad (\text{F13})$$

Substituting Eq. (F13) into Eq. (F6), we obtain

$$\begin{aligned} \Delta\langle U_{ij} \rangle_{\text{aniso}} &= -\frac{3\mu_0}{4\pi r_{ij}^3} \int_{-\infty}^{\infty} C_k(\Delta u) D_m(\Delta u) d\Delta u, \\ D_m(\Delta u) &= C_{m,y}(\Delta u) - C_{m,x}(\Delta u), \end{aligned} \quad (\text{F14})$$

which corresponds to Eqs. (15) and (17).

As written in the main text, changing the variable from the time lag Δu to the phase lag $\Delta\xi$ in Eq. (15) makes the $\tau\omega$ dependence of $\Delta\langle U_{ij} \rangle_{\text{aniso}}$ explicit. Letting $\Delta\xi = \omega\Delta u$, $\xi = \omega t$, and $\omega T_{\text{cyc}} = 2\phi = 4\pi$ (under a regular ARMF), we can rewrite Eq. (F14) as

$$\Delta\langle U_{ij} \rangle_{\text{aniso}} = -\frac{3\mu_0}{4\pi r_{ij}^3} \int_{-\infty}^{\infty} C_k(\tau\omega, \Delta\xi) D_m(\Delta\xi) d\Delta\xi, \quad (\text{F15})$$

where

$$\begin{aligned} C_k(\tau\omega, \Delta\xi) &= \frac{1}{2\tau\omega} \exp\left(-\frac{|\Delta\xi|}{\tau\omega}\right), \\ D_m(\Delta\xi) &= C_{m,y}(\Delta\xi) - C_{m,x}(\Delta\xi), \end{aligned}$$

$$\begin{aligned} C_{m,\sigma}(\Delta\xi) &= \frac{1}{4\pi} \int_0^{4\pi} m_{\text{eq},\sigma}(\xi) m_{\text{eq},\sigma}(\xi - \Delta\xi) d\xi, \\ \sigma &= x, y. \end{aligned}$$

We use the following two key properties: $C_k(\tau\omega, \Delta\xi)$ is even in $\Delta\xi$, and $D_m(\Delta\xi)$ is even and 4π periodic (for a regular ARMF with $\omega T_{\text{cyc}} = 4\pi$),

$$\begin{aligned} C_k(\tau\omega, -\Delta\xi) &= C_k(\tau\omega, \Delta\xi), \\ D_m(-\Delta\xi) &= D_m(\Delta\xi), \\ D_m(\Delta\xi + 4\pi) &= D_m(\Delta\xi). \end{aligned} \quad (\text{F16})$$

By evenness of $C_k(\tau\omega, \Delta\xi)$ and $D_m(\Delta\xi)$, the integrand is even in $\Delta\xi$, so the integral over $\Delta\xi \in (-\infty, \infty)$ is twice the integral over $\Delta\xi \in [0, \infty)$. We then partition the positive half-line into intervals of length 4π by writing $\Delta\xi = \Delta\xi' + 4\pi n$, with $\Delta\xi' \in [0, 4\pi)$, where n is a non-negative integer. For $\Delta\xi \geq 0$, the kernel autocorrelation can be written as

$$\begin{aligned} C_k(\tau\omega, \Delta\xi' + 4\pi n) &= \frac{1}{2\tau\omega} \exp\left(-\frac{\Delta\xi' + 4\pi n}{\tau\omega}\right) \\ &= \exp\left(-\frac{4\pi n}{\tau\omega}\right) C_k(\tau\omega, \Delta\xi'). \end{aligned} \quad (\text{F17})$$

Using this result together with the 4π periodicity of $D_m(\Delta\xi)$, we obtain

$$\begin{aligned} &\int_0^\infty C_k(\tau\omega, \Delta\xi) D_m(\Delta\xi) d\Delta\xi \\ &= \left(\int_0^{4\pi} C_k(\tau\omega, \Delta\xi) D_m(\Delta\xi) d\Delta\xi \right) \sum_{n=0}^\infty \exp\left(-\frac{4\pi n}{\tau\omega}\right). \end{aligned} \quad (\text{F18})$$

Evaluating the geometric series and using $C_k(\tau\omega, \Delta\xi) = (2\tau\omega)^{-1} \exp(-\Delta\xi/(\tau\omega))$ on $\Delta\xi \in [0, 4\pi)$, we arrive at

$$\begin{aligned} &\int_{-\infty}^\infty C_k(\tau\omega, \Delta\xi) D_m(\Delta\xi) d\Delta\xi \\ &= \frac{1}{\tau\omega(1 - \exp(-\frac{4\pi}{\tau\omega}))} \int_0^{4\pi} \exp\left(-\frac{\Delta\xi}{\tau\omega}\right) D_m(\Delta\xi) d\Delta\xi. \end{aligned} \quad (\text{F19})$$

Substituting this result into Eq. (F15) yields Eq. (19).

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