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Discontinuous Molecular Dynamics Simulations in an External Field: Application in Two-Dimensions

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We introduce a stochastic method for simulating the effect of an external magnetic field on coarse-grained models of magnetic colloids for use in discontinuous molecular dynamics (DMD) simulations. Our method for simulating an external field is illustrated with a coarse-grained model for magnetic squares in two-dimensions. Square-shaped particles are represented as four discs bonded together in a 2x2 lattice configuration to create a hard colloidal geometry. Two opposite charges are embedded within the square to mimic the magnetic interactions between particles. The method for simulating an external field stochastically during DMD simulations operates by applying impulses randomly to the charges embedded within each square particle. When one square experiences an interaction with the field, each embedded charge within the square is assigned a new momentum with a specific magnitude and orientation. The magnitude of this momentum is equal to the average of a Maxwell-Boltzmann distribution at the simulation temperature. The orientation of the momentum depends on the charge, either positive or negative, and points either in the same or opposite direction as the field, respectively. The strength of the external field is determined by the average frequency at which the particles experience interactions with the field. The relationship between the stochastic frequency of the field and the field strength is derived from Newton’s equation of motion. DMD simulations are performed for large systems of magnetic square particles at various temperatures and external field strengths. The simulation temperature is maintained constant with an Andersen thermostat, while the external field is simulated stochastically, as described above. We find that our simulation techniques reproduce a net system magnetization in close agreement with the two-dimensional equivalent of the Langevin function, while maintaining the simulation temperature constant.

I. INTRODUCTION

Systems of colloidal particles self-assemble into hierarchical structures that span several length scales and form the basis of many different types of materials [1]. New types of colloid-based functional materials with tunable interactions are being synthesized via bottom-up design, wherein particle-particle interactions are engineered to encourage the formation of desired assemblies [2]. One way to direct the assembly of a colloidal system into an organized structure is to apply an external magnetic field. Magnetic fields polarize certain susceptible materials (e.g. aluminum, cobalt), and then orient particles made from or containing those materials in the field’s direction [3]. Furthermore, external fields can be programmed to change with time (e.g. field rotation or toggling), causing systems of particles to shift into unique, out-of-equilibrium assemblies [4, 5]. A challenge in designing these types of functional materials is to predict the relationship between the particle-particle interactions and their macroscopic properties. Computer simulations are often utilized as a low-cost guide to explore the assembling behavior of novel colloidal systems [6–8].

We are interested in applying computational models that can predict the macroscopic properties that arise from the self-assembly of ferromagnetic particles with anisotropic features at the colloidal scale. Particles made from or containing ferromagnetic materials retain their magnetic polarization, or magnetization, even af-

ter a polarizing magnetic field is decreased or turned off. Examples of colloidal ferromagnetic particles include microscopic patchy colloids, in which the particle surface is coated in a ferromagnetic material, and metallic nanorods and nanocubes, which are synthesized from ferromagnetic materials [9–12]. Systems of ferromagnetic particles at the colloidal scale are referred to as ferrofluids and have superparamagnetic properties [13, 14]. Superparamagnetic systems display reversible polarization, which means that they have a high degree of magnetization in the presence of an external field, but that the magnetization dissipates when the external field is removed. Superparamagnetic materials have found applications in areas like magnetic actuators [15–17], magnetic separation [18, 19], and drug delivery systems [20, 21].

Here, we present a stochastic method for simulating the effect of an external field on a system of magnetic colloidal squares during coarse-grained molecular dynamics simulations (Figure 1.a). Simulations of our model system are performed using an event-driven version of the standard molecular dynamics procedure called Discontinuous Molecular Dynamics (DMD) [22, 23]. To the best of our knowledge, there does not yet exist a method for simulating an external field that is applicable to DMD. We show that our method for simulating an external field stochastically is thermodynamically consistent: when DMD simulations in a stochastic external field are coupled with the Andersen thermostat, the simulation temperature is constant and the super-

paramagnetic behavior of the system in the presence of the stochastically-applied external field is very close to the behavior predicted by Boltzmann statistics in two dimensions, as originally explored by Langevin [24, 25].

II. COARSE-GRAINED MODEL FOR MAGNETIC PARTICLES

Our model consists of a system of magnetic particles in two dimensions. The particles have square-like geometries and contain embedded magnetic dipoles along one edge as shown in Figure 1.a. The excluded volume interactions of the squares are modeled as four hard discs of diameter σ in a 2×2 lattice configuration. The interaction between two bonded hard discs is modelled using Bellemans' approach as

$$U_B^*(r_{ij}^*, d_B^*) = \begin{cases} \infty & \text{if } r_{ij}^* \leq d_B^* - \delta \\ 0 & \text{if } d_B^* - \delta < r_{ij}^* < d_B^* + \delta \\ \infty & \text{if } r_{ij}^* \geq d_B^* + \delta \end{cases} \quad (1)$$

where r_{ij}^* is the reduced distance between two bonded discs, d_B^* is the reduced diameter of the bond, and δ is a constant that defines how tightly two discs are bonded together [26]. For our models, all distances are reduced by the hard disc diameter σ . In total, five bonds are used to maintain the four hard discs making the magnetic square. Four bonds with diameter $d_B^* = 1$ constrain the hard discs to their adjacent neighboring discs. One bond with diameter $d_B^* = \sqrt{2}$ constrains one set of hard discs located diagonally across from one another in the square formation. For our simulations, Bellemans' constant is

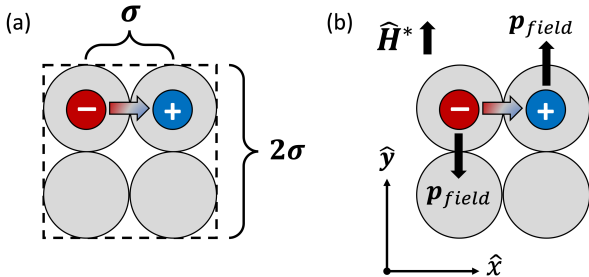


FIG. 1. (a) *Coarse-grained model for magnetic squares.* Grey circles represent hard discs, and red and blue circles represent the negative and positive charges, respectively. The black, dashed line outlines the square shape formed by the hard discs. (b) *Method for simulating an external field stochastically.* When a magnetic square experiences a field collision, each of the charges is assigned a new momentum $\mathbf{p}_{field}$. For positive charges, the field impulse points in the same direction as the field; for negative charges, the field impulse points in the opposite direction to the field. The coordinate system is shown in the bottom left corner; the field orientation $\hat{\mathbf{H}}^*$ is in the positive y-axis direction.

set to $\delta = 0.015$. The hard neutrally-charged discs on each square interact with other hard neutrally-charged discs on nearby squares via an infinitely-repulsive hard-disc potential, modeled as

$$U_{HS}^*(r_{ij}^*) = \begin{cases} 0 & \text{if } r_{ij}^* > 1 \\ \infty & \text{if } r_{ij}^* \leq 1 \end{cases} \quad (2)$$

where r_{ij}^* is the reduced distance between two unbonded discs [27].

Models for magnetic particles can be classified by how the dipolar magnetic interactions are modelled: either as a point dipole or as an extended dipole. In the case of the former, the magnetic interactions are represented by the point dipole equation, which is applicable to magnetic particle in which the distance of charge separation is small relative to the distance separation between two magnetic dipoles [28]. In the latter case, the magnetic interactions are represented as two opposite charges; this is applicable to magnetic particles in which the degree of charge separation is on the same length scale as the distance of separation between two magnetic dipoles [29, 30]. In our models, the magnetic dipole is modeled as an extended dipole, such that two embedded ‘charges’, equal in strength but opposite in magnitude, are separated by a distance of σ . The dipole points from the negative to the positive charge and is oriented clockwise relative to the square’s center of mass. The charges embedded in each square interact with charges in nearby squares via a short-range discontinuous potential. Opposite charges interact with an attractive, square-well potential U_{SW}^* , and like charges interact with a repulsive, square-shoulder potential U_{SS}^* . The square-well potential U_{SW}^* is described as

$$U_{SW}^*(r_{ij}^*) = \begin{cases} -1 & \text{if } r_{ij}^* < 1.15 \\ -0.3 & \text{if } r_{ij}^* < \sqrt{2} \\ 0 & \text{if } r_{ij}^* \geq \sqrt{2} \end{cases} \quad (3)$$

where r_{ij}^* is the distance between two opposite charges. For our models, all interactions are reduced by ϵ which has units of energy and parameterizes the depth of the square-well potential U_{SW}^* , thereby representing the strength of the magnetic interaction between two squares. The attractive (square-well) and repulsive (square-shoulder) potentials are related according to $U_{SS}^* = -U_{SW}^*$; the widths are the same, and the depths are opposite. The depths of the square well potential U_{SW}^* are selected to mimic a continuous Yukawa potential, also known as a screened columbic potential [31]. The reduced magnitude of each charge q^* is calculated as $q^* = q(4\pi\epsilon_0\epsilon\sigma)^{-\frac{1}{2}}$, where ϵ_0 is the permittivity of free space. The reduced dipole moment μ^* is $\mu^* = q^*l^*$, where l^* is the reduced distance of separation between two embedded charges. This model for magnetic squares was used previously by us to explore the self-assembly of

colloidal magnetic squares in the absence of an external field [31]. More generally, this method of coarse-graining magnetic colloidal particles has been used by our group to explore the self-assembly of other magnetic colloidal systems, such as magnetic rods and discs [32–35].

III. STOCHASTIC METHOD FOR SIMULATING AN EXTERNAL FIELD AT CONSTANT TEMPERATURE

Discontinuous molecular dynamics simulations are performed for systems containing the model magnetic square particles confined to a two-dimensional plane with periodic boundaries. The system is described by an area fraction, ϕ , calculated as the ratio of the area excluded by all magnetic squares in the system to the total area of the simulation plane. The Andersen thermostat maintains the simulation temperature constant by mimicking stochastic interactions with an external heat bath [24]. In this method, randomly chosen magnetic squares, selected according to a Poisson distribution, experience stochastic impulses with the thermostat at a frequency $\nu_{thermostat}$ [36]. When a square is selected to experience a random thermostat event, each disc within the square is assigned a unique momentum according to a two-dimensional Maxwell-Boltzmann distribution that corresponds to the reduced simulation temperature defined to be $T^* = k_B T / \varepsilon$, where k_B is the Boltzmann constant. For our simulations, the frequency with which squares are randomly selected to experience collisions with the thermostat is set to $\nu_{thermostat} = 0.2 t^{*-1} \text{square particle}^{-1}$, where t^* is the simulation time scale defined in reduced seconds as $t^* = t \sqrt{\varepsilon / \sigma^2 \cdot m}$ (σ and m are the diameter and mass of one hard disc, respectively). For any simulation, the simulation temperature, $T_{measured}$, should match the assigned temperature such that $T_{measured} = T^*$, which is calculated as $T_{measured} = \langle \mathbf{p}_i^2 \rangle / 2m$, an ensemble average $\langle \rangle$ of the system's kinetic energy in terms of each particle's momentum $\mathbf{p}_i$.

Next, we introduce a method for simulating the effect of an external field on our model system during DMD simulations. The reduced external field $\mathbf{H}^*$ is defined by a scalar magnitude, H^* , and an orientation, $\hat{\mathbf{H}}^*$, such that $\mathbf{H}^* = H^* \hat{\mathbf{H}}^*$. The reduced external field strength H^* is related to the real external field strength H according to $H^* = (\sigma^3 / \varepsilon)^{1/2} H$. The effect of the field on the magnetic particles is simulated by applying stochastic impulses to the charges embedded in the squares in such a way that the magnetic particles tend to become oriented in the direction of the field, as illustrated in Figure 1.b. This approach is similar to the way that the Andersen thermostat procedure maintains the simulation temperature. Unlike the Andersen thermostat, however, the simulated field is only applied to pairs of charges that are embedded within the colloidal square.

When one magnetic square experiences a field collision, both charges embedded within that square are assigned new momenta, $\mathbf{p}_{field}$, which represents an impulse applied to the center of that charge. The momentum imparted by the field to any charge is described by a scalar magnitude, p_{field} , and an orientation, $\hat{\mathbf{p}}_{field}$, such that $\mathbf{p}_{field} = p_{field} \hat{\mathbf{p}}_{field}$. The magnitude of the momentum imparted by the field, p_{field} , is constant and equal to the average of a two-dimensional Maxwell-Boltzmann distribution at the simulation temperature T^* , such that $p_{field} = \sqrt{2T^*}$. The orientation of the momentum imparted by the field, $\hat{\mathbf{p}}_{field}$, is related to the field orientation $\hat{\mathbf{H}}^*$ and the embedded charge q_i^* , such that $\hat{\mathbf{p}}_{field} = q_i^* \hat{\mathbf{H}}^*$. Consequently, the momentum assigned to charges is

$$\mathbf{p}_{field} = q_i^* \sqrt{2T^*} \hat{\mathbf{H}}^* \quad (4)$$

The strength of the external field, H^* , is controlled by adjusting the frequency at which magnetic squares experience interactions with the field, ν_{field} . We derive this relationship from Newton's equation of motion,

$$\mathbf{F}_i = \frac{d\mathbf{p}_i}{dt} \quad (5)$$

which relates the force, $\mathbf{F}_i$, that a charge experiences to the first time derivative of the particle's momentum, $d\mathbf{p}_i/dt$. For our models, the force that a charged particle experiences in a field is $\mathbf{F}_i = q_i^* \mathbf{H}^*$. Furthermore, we define the first time derivative of the particle's moment as $d\mathbf{p}_i/dt = \nu_{field} \cdot \mathbf{p}_{field} = \nu_{field} q_i^* \sqrt{2T^*} \hat{\mathbf{H}}^*$ (from Equation (4)), which is the impulse imparted by the field one over charge over the average period between stochastic collisions from the field (ν_{field}^{-1}). Consequently, Newton's equation of motion becomes

$$q_i^* \mathbf{H}^* = \nu_{field} q_i^* \sqrt{2T^*} \hat{\mathbf{H}}^*. \quad (6)$$

By simplifying Equation (6), is the relationship the frequency of stochastic interactions with the external field ν_{field} and the magnitude of the external field H^* becomes

$$H^* = \nu_{field} \sqrt{2T^*} \quad (7)$$

According to Equation (7), the frequency at which magnetic particles experience interactions with the external field is a function not only of the external field strength, H^* , but also of the assigned simulation temperature, T^* . The method for randomly selecting particles to experience interactions with the field is identical to that of the Andersen thermostat whereby randomly-chosen magnetic squares, selected according to a Poisson distribution, experience stochastic impulses with the field at an average frequency of ν_{field} .

During any simulation the measured magnetization of the system, $M_{measured}$, should match the predicted value, $M_{expected}$. Classical magnetic theory estimates the

macroscopic magnetization of a large system of magnetic dipole moments in terms of the probability, $Prob$, that a single magnetic dipole moment will adopt an orientation θ relative to an external field. $Prob$ is calculated as

$$Prob(\theta, X) = e^{X \cos \theta} \quad (8)$$

where $X \equiv \mu^* H^* / T^*$ and represents the normalized field strength, or the magnetic energy of one magnetic dipole moment in an external field, relative to the system temperature.

The Boltzmann relationship (Equation (8)) is the basis for predicting the magnetic response of a dilute system of non-interacting dipoles in two-dimensions to an external magnetic field, $M_{expected}$. The normalized probability distribution $f(\theta, X)$ that a magnetic dipole moment adopts an angle θ at a normalized field strength X is

$$f(\theta, X) = \frac{Prob(\theta, X)}{\int_{-\pi}^{\pi} Prob(\theta, X) d\theta} = \frac{e^{X \cos \theta}}{2\pi I_0(X)} \quad (9)$$

where I_0 is a modified Bessel function of order zero. Equation (9) is also known as the circular von Mises distribution; it predicts the relative likelihood of finding a magnetic dipole at any angle θ relative to an external field [37]. The expected magnetization, $M_{expected}$, is the first moment of $f(\theta, X)$, and is calculated from Equation (9) as

$$M_{expected}(X) = \int_{-\pi}^{\pi} \cos \theta f(\theta, X) d\theta = \frac{I_1(X)}{I_0(X)} \quad (10)$$

where I_1 is a modified Bessel function of the first order. The expected magnetization in two dimensions, $M_{expected}$ was originally predicted by Jiles, *et al.* [38]. For any simulation, the expected magnetization, $M_{expected}$, should match the measured system magnetization, $M_{measured}$, corresponding to the normalized field strength, X , such that $M_{measured} = M_{expected}(X)$. The measured system magnetization, $M_{measured}$, is calculated during simulations as the magnetic order parameter $M_{measured} = \langle \hat{\mu}_i \cdot \hat{H}^* \rangle$.

IV. RESULTS

Simulations are performed for $N_{squares} = 1,024$ magnetic squares at a dilute area fraction of $\phi = 0.03$ in the presence of a stochastically-simulated external field with strength H^* and orientation in the positive y-direction, i.e. $\hat{H}^* = \hat{y}$, as shown in Figure 1.b. The simulation temperature, T^* , is maintained using the Andersen thermostat. The simulations start from a random configuration and are performed for 400 million events, where one simulation event is either a collision between two particles, or a stochastic thermostat or field event. After the 400 million events have been completed, the system is considered to be equilibrated and the properties $T_{measured}$ and

$M_{measured}$ are calculated every 100,000 events for an additional 50 million collision events. DMD simulations are performed at 17 unique temperatures T^* ranging from 0.20 (a relatively low temperature for our model) to 1.0 (an arbitrarily high temperature), and for a total of 251 unique normalized field strengths, X , ranging from 0.00 (field is off) to 5.0 (a high external field strength relative to the system temperature). The external field strength H^* assigned to a simulation is calculated from the normalized external field strength, X , according to

$$H^* = \frac{X \cdot T^*}{\mu^*} \quad (11)$$

Simulations are performed for each of the 4,267 unique combinations of assigned temperature and normalized external field strength in replicates of 5, resulting in 21,335 unique simulations. The equilibrium properties at each unique combination of assigned temperature T^* and normalized field strength X are determined by averaging the simulation properties returned by each of the 5 replicate simulations.

The measured system temperature, $T_{measured}$, and magnetization, $M_{measured}$, for each of the conditions described above are plotted against the normalized external field strength X for various temperatures T^* in Figure 2.a and Figure 2.b, respectively. In Figure 2.a, we observe that $T_{measured}$ remains at the assigned value regardless of the normalized external field strength, X , which confirms that the stochastic method for simulating the external field does not cause the system temperature to deviate from the assigned value, T^* . In Figure 2.b, we find that the magnetization $M_{measured}$ for simulations performed at different temperatures (colored lines) have very similar behavior regardless of the temperature that is assigned to the simulation. When compared to the magnetization that is predicted by Boltzmann statistics, $M_{expected}$ (dashed line calculated from Equation (10)), the measured system magnetization, $M_{measured}$ (colored lines), is very close to the values predicted by theory. The asymptotic behavior of Equation (10) is such that as $X \rightarrow \infty$, $M_{measured}(X) \rightarrow 1$. The magnetization calculated from our simulated system has approximately the same asymptotic behavior. However, we do find that the system is slightly more magnetized by the field than is predicted by theory, especially at high normalized external field strengths X .

In Figure 3, we show that the difference between net magnetization predicted by theory and the magnetic response of our simulated system in response to the external field observed in Figure 2.b is a consequence of the magnetic particles being slightly more likely to point in the field direction than predicted by theory. We do this by comparing the expected angular distribution of a magnetic dipole in an external field $f(\theta, X)$ (Equation 9) to the average angular distribution of magnetic square particles with respect to the orientation of the external

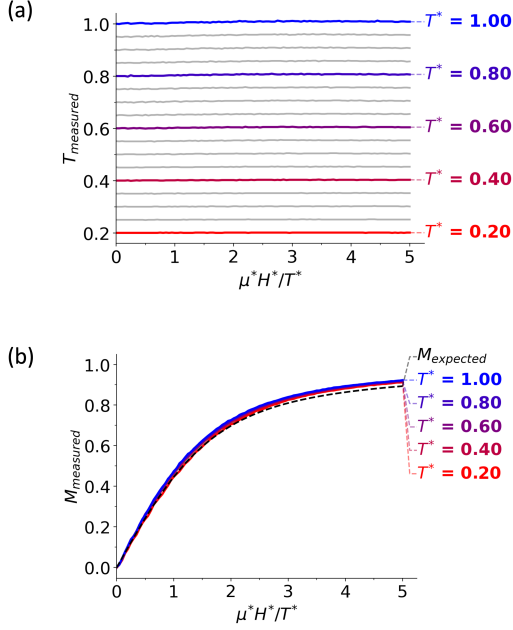


FIG. 2. Measured system temperature T_{measured} (a) and system magnetization M_{measured} (b) against normalized external field strength μ^*H^*/T^* , or X , for various assigned temperatures T^* . Highlighted lines are isotherms labeled according to their assigned temperature, where the color of the line indicates the value associated with the isotherm. Gray lines are isotherms performed at simulated temperatures that are not labeled. The black, dashed line in (b) is the expected system magnetization predicted by classical magnetic theory, calculated according to Equation (10).

field during DMD simulations. The angular distributions, which are shown as a histogram in Figure 3, were calculated once the simulation had equilibrated in simulations performed at $T^* = 0.60$ and at selected normalized external field strengths X . We find that at relatively low normalized external field strengths, X , the measured angular distribution is very similar to that predicted by theory (dashed line in Figure 2, calculated from Equation 9). However, as the strength of the external field increases, the simulation distribution deviates from the expected distribution. At relatively high external field strengths, systems of magnetic particles are more likely to point in the direction of the field ($\theta = 0$) than is predicted by theory. The deviation between the expected distribution and the simulation distribution provides an explanation for the deviation between the expected M_{expected} and the measured M_{measured} system magnetization, which occurs at high values of the normalized external field strengths and is observed in Figure 2.b. Since the magnetic particles are slightly more oriented in the direction of the field than theory predicts, the overall magnetization of the system is slightly greater than that predicted by theory.

Finally, in Figure 4, the measured system magneti-

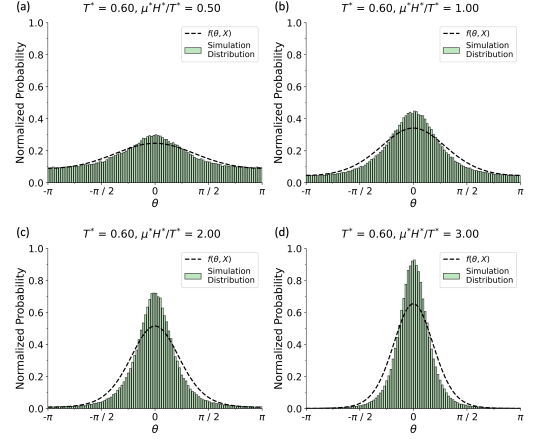


FIG. 3. Angular distribution of squares in a stochastic external field. Comparison of expected angular distribution of magnetic dipoles in an external field (black, dashed line) to simulation data (green bars) for different normalized external field strengths X and at a constant temperature of $T^* = 0.60$.

zation, M_{measured} , is plotted against the external field strength H^* . We find that the system's magnetization in response to the external field strength, also known as the paramagnetic susceptibility of the system, χ , is inversely proportional to the system temperature. At low simulated temperatures (e.g. $T^* = 0.20$), the system's magnetization is significantly greater than the magnetization at higher simulated temperatures (e.g. $T^* = 1.00$). This behavior is characteristic of the magnetic behavior of superparamagnetic systems, where the paramagnetic susceptibility is inversely related to the system temperature [39].

V. CONCLUSIONS

We have introduced a stochastic method for simulating the effect of an external field, which we apply to coarse-grained models for magnetic colloidal particles during discontinuous molecular dynamics simulations. Our method mimics interactions between a magnetic particle and an external field by exerting random impulses on magnetic particles during DMD simulations. The magnitude of the impulses applied to magnetic particles is related to the simulation temperature, while the frequency of the impulses controls the strength of the applied field. We apply this method for simulating an external field to a coarse-grained model of magnetic square colloids. When calculating the magnetic polarization of the system versus the normalized external field strength, we find that the system's magnetization in response to the field is very close to that predicted by Boltzmann statistics and that the simulation temperature does not deviate from the assigned value. Our method for simu-

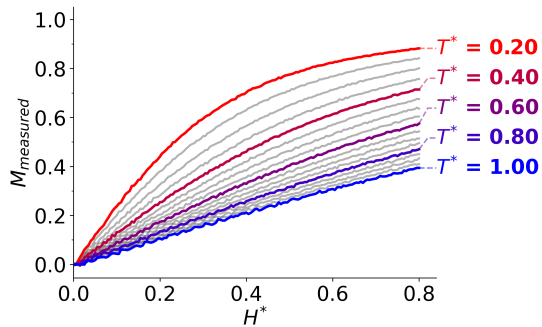


FIG. 4. Measured system magnetization $M_{measured}$ against external field strength H^* for various assigned temperatures T^* . Highlighted lines are isotherms labeled according to their assigned temperature, where the color of the line indicates the value associated with the isotherm. Gray lines are isotherms performed at simulated temperatures that are not labeled.

lating an external field is generalized in such a way that it can be applied to other models for magnetic particles in which the magnetic interactions are modeled as extended dipoles, including those that exist in three-dimensions. Models of this type have been used extensively to explore the self-assembly of magnetic particles with various types of anisotropy in the absence of an external field. Our method for simulating an external field stochastically can be used to extend the applicability of these models to simulations in the presence of an external field. Here, we apply our simulation techniques during DMD simulations; however, our simulation methods have been generalized in such a way that they could be applied during standard molecular dynamics simulations as well.

While we can conclude that the stochastic method presented here sufficiently reproduces the behavior predicted by theory, there does exist a small error between the magnetization expected by Boltzmann statistics and that reproduced by the model system, especially at high simulated field strengths (see Figure 2.b and Figure 3). Due to the coarse-grained nature of our model, it is difficult to deduce whether the deviation in the system's response to the stochastic field is due to the model, which is coarse-grained, or to the stochastic simulation method, which is completely new. It may be that the slight variation in the length of the dipole in our model (which is represented as opposite charges tethered together by a flexible bond) introduces differences between simulation results and theory, which models the dipole length as constant. An alternative source of error could be our method for simulating a force field stochastically, which applies constant-magnitude and constant-direction impulses to the charged particles by the stochastic force field. By allowing some variance (in a thermodynamically consistent way) to the orientation or magnitude of the impulses that are applied to charges during simulations,

the over magnetization observed during simulations, shown in Figure 3, might be reduced to match the magnetization predicted by theory. Our technique for representing a force-field through stochastic interactions is new, so further investigation of this stochastic method would prove insightful, especially for those seeking to develop coarse-grained force-fields. Our method for coarse-graining constant forces via discontinuous, or stochastic, forces has not been used before to the best of our knowledge. It would be interesting to explore what other types of force fields might be represented equally well as stochastic interactions, or vice versa.

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