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Dynamic symmetry loss of high-frequency hysteresis loops in single-domain particles with uniaxial anisotropy

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ABSTRACT

Understanding how magnetic materials respond to rapidly varying magnetic fields, as in dynamic hysteresis loops, constitutes a complex and physically interesting problem. But in order to accomplish a thorough investigation, one must necessarily consider the effects of thermal fluctuations. Albeit being present in all real systems, these are seldom included in numerical studies. The notable exceptions are the Ising systems, which have been extensively studied in the past, but describe only one of the many mechanisms of magnetization reversal known to occur. In this paper we employ the Stochastic Landau–Lifshitz formalism to study high-frequency hysteresis loops of single-domain particles with uniaxial anisotropy at an arbitrary temperature. We show that in certain conditions the magnetic response may become predominantly out-of-phase and the loops may undergo a dynamic symmetry loss. This is found to be a direct consequence of the competing responses due to the thermal fluctuations and the gyroscopic motion of the magnetization. We have also found the magnetic behavior to be exceedingly sensitive to temperature variations, not only within the superparamagnetic–ferromagnetic transition range usually considered, but specially at even lower temperatures, where the bulk of interesting phenomena is seen to take place.

1. Introduction

Dynamic hysteresis has been the subject of intensive research in the past, particularly in the context of Ising ferromagnets, in which it has been shown that the system may undergo a first-order phase transition resulting in asymmetrical loops [1]. Poperechny et al. [2] recently pointed to the possibility of a similar transition taking place in single-domain particles. In this system, the exchange interactions dominate over the others inhibiting the formation of magnetic domains [3]. This forces the magnetization to behave in unison, analogously to a magnetic dipole moment of fixed magnitude undergoing gyroscopic motion. It is thus remarkable that the same transition should take place in such system. For, while in Ising ferromagnets the magnetization reversal occurs through domain nucleation, in single-domain particles the only allowable mode is through coherent rotation. The universal character of a same phenomena arising from completely different mechanisms renders a numerical investigation of this problem pertinent.

A striking difference between Ising systems and single-domain particles is the enhanced thermal instability of the latter [4,5]. It is well known that their magnetic properties change drastically from the high-temperature superparamagnetic state, where the thermal fluctuations hinder any magnetic order, to the low-temperature, magnetically stable state. In slow measurements, reducing the temperature even further has only minor effects on the magnetic response [6]. In contrast, we will show that in high-frequency hysteresis loops [7–9] most interesting effects take place at temperatures bellow the ferromagnetic order. Thus, albeit the similarities with Ising systems, single-domain particles have very unique properties as a consequence on the non-linear competition between the thermal fluctuations and the gyroscopic response time. We note in passing that the situations here examined are within reach of current experimental techniques and moreover, may also be of practical importance, for instance in the context of magnetic memories which demand increasingly faster operation speeds.

2. Computational details

In order to study the high-frequency hysteresis loops of single-domain particles we numerically solved the Stochastic Landau–Lifshitz (SLL) equation [2,10–16], which may be compactly written using reduced variables as

$$\frac{d\boldsymbol{m}}{dt} = -\frac{1}{\alpha}\boldsymbol{m}\times(\boldsymbol{h}+\boldsymbol{h}_{th}) - \boldsymbol{m}\times[\boldsymbol{m}\times(\boldsymbol{h}+\boldsymbol{h}_{th})], \tag{1}$$

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where $\boldsymbol{m}$ is the magnetization unit vector and α is the damping constant. Time is already assumed normalized in units of

$$\tau_0 = \frac{1+\alpha^2}{\alpha\gamma_0 H_A},$$

where $\gamma_0 = 2.211 \times 10^5$ m/(A s) is the gyromagnetic ratio of the electron and $H_A = 2K_1/M_s$ is the zero temperature coercivity [3] (K_1 is the first uniaxial anisotropy constant and M_s is the saturation magnetization of the material). In Eq. (1) the deterministic field ($\boldsymbol{h}$) comprises the anisotropy field and the external field ($\boldsymbol{h}_{ap}$), assumed to have a simple harmonic form. Both are normalized in units of H_A and assumed to be parallel to the z direction (unit vector $\hat{\boldsymbol{e}}_z$). Thus,

$$\boldsymbol{h}(t) = (h_0 \cos(\omega t) + m_z)\hat{\boldsymbol{e}}_z. \tag{2}$$

The thermal field ($\boldsymbol{h}_{th}$) is taken as a Gaussian white noise with first two moments

$$\langle \boldsymbol{h}_{th}(t) \rangle = 0,$$

$$\langle h_{th_i}(t) h_{th_j}(s) \rangle = \frac{\alpha^2}{(1+\alpha^2)} \frac{1}{\sigma} \delta_{i,j} \delta(t-s),$$

where the Kronecker and Dirac deltas describe the absence of spatial and temporal correlations respectively. Here σ is defined as the ratio between the anisotropy energy barrier and the thermal energy:

$$\sigma = \frac{K_1 V}{k_B T}. \tag{3}$$

We numerically solved the stochastic differential equation (1) using Heun's scheme [10,14,17] with a fixed step size of 10^{-4} and damping $\alpha = 0.1$. For each condition we computed 300 stochastic trajectories for 20 cycles of the external field. In order to minimize transient effects we set for half of the trajectories to have $\boldsymbol{m}(t=0) = \hat{\boldsymbol{e}}_z$ and for the other half to have $\boldsymbol{m}(t=0) = -\hat{\boldsymbol{e}}_z$. The first five cycles were then discarded and the resulting ones averaged so as to obtain the average magnetization in the direction of the field, denoted simply as m.

3. Results

The complex many-body character of Ising systems inhibits a simple explanation of the dynamic symmetry loss. Fortunately, this is not the case for single-domain particles in which this effect may be qualitative understood, at least to some extend, by the following arguments. Suppose that $h_0 = 1$ in Eq. (2). In the context of the energy landscape of Stoner and Wolfarth [3], if the magnetization starts with $\boldsymbol{m} = \hat{e}_z$, it will remain so until $h_{ap} = -1$, when it suddenly flips to the opposite pole. The presence of thermal fluctuations lifts this restriction, allowing the particle to flip when $h_{ap} < 1$. In slow hysteresis loops this usually translates into a reduction of the coercivity with increasing temperature; that is, the reversal occur for $t < \pi/\omega$. But note that this does not necessarily has to be so and it may just as well happen when $t > \pi/\omega$. As will be shown, this is the precise condition for the dynamic symmetry loss, which results in the majority of the macrospins becoming out-of-phase with the magnetic field. This remarkable effect is due entirely to the thermal fluctuations; it does not exist at zero temperature. It occurs at intermediate-to-high frequencies and low temperatures, in situations where the thermal fluctuations, albeit being considerably suppressed, aid the external field in promoting reversals that would otherwise be inhibited by the gyroscopic response time of the magnetization. Following this discussion, we define the "coercive time" (t_c) as the first instant of time during a cycle where $m(t_c) \equiv 0$.

In Fig. 1 we have compiled hysteresis loops for different values of ω (rows) and σ (columns), and for three amplitudes of the external field; namely $h_0 = 1.0$, 1.5 and 2.0. We also denote the positions of m at the beginning and midway through the cycle ($m(t=\pi/\omega)$), by red and blue dots respectively. Note that $\sigma \propto 1/T$, i.e. the dependence is asymptotic. Several qualitative conclusions may be drawn from this image. By following the colored dots it is possible to see that between $\sigma = 1$ and $\sigma \simeq 10$ the height of the loops increases, signifying a reduction of thermal jumps. This region corresponds to the transition from a superparamagnetic to a ferromagnetic state. Upon further increasing σ, the dots tend toward the abscissa and after crossing it, the loops tilt to the left, becoming asymmetrical (t_c has surpassed π/ω). Continuing even further, at very low temperatures, the inter-well jumps become quickly suppressed and the loops shrink to zero. On the other

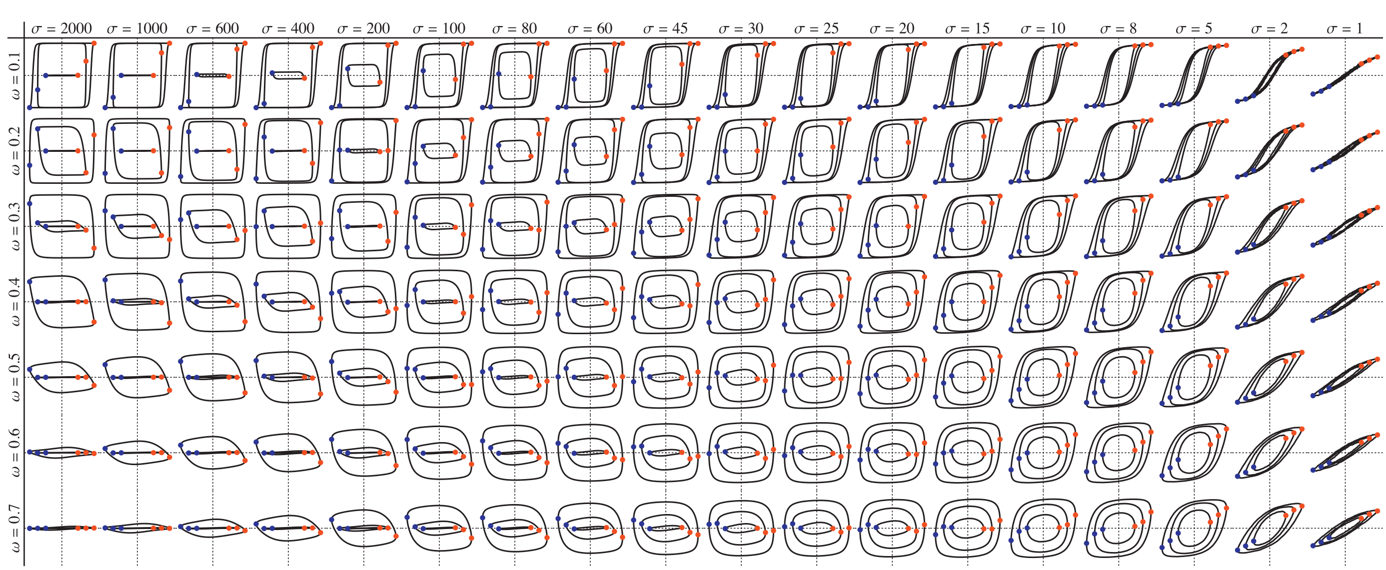

Fig. 1. Hysteresis loops for different frequencies (rows) and temperatures (columns). Each image shows three loops corresponding to $h_0 = 1.0$, 1.5 and 2.0 (from inner to outermost). Red and blue dots denote the positions at the beginning and midway through the cycle ($m(t=0)$ and $m(t=\pi/\omega)$). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

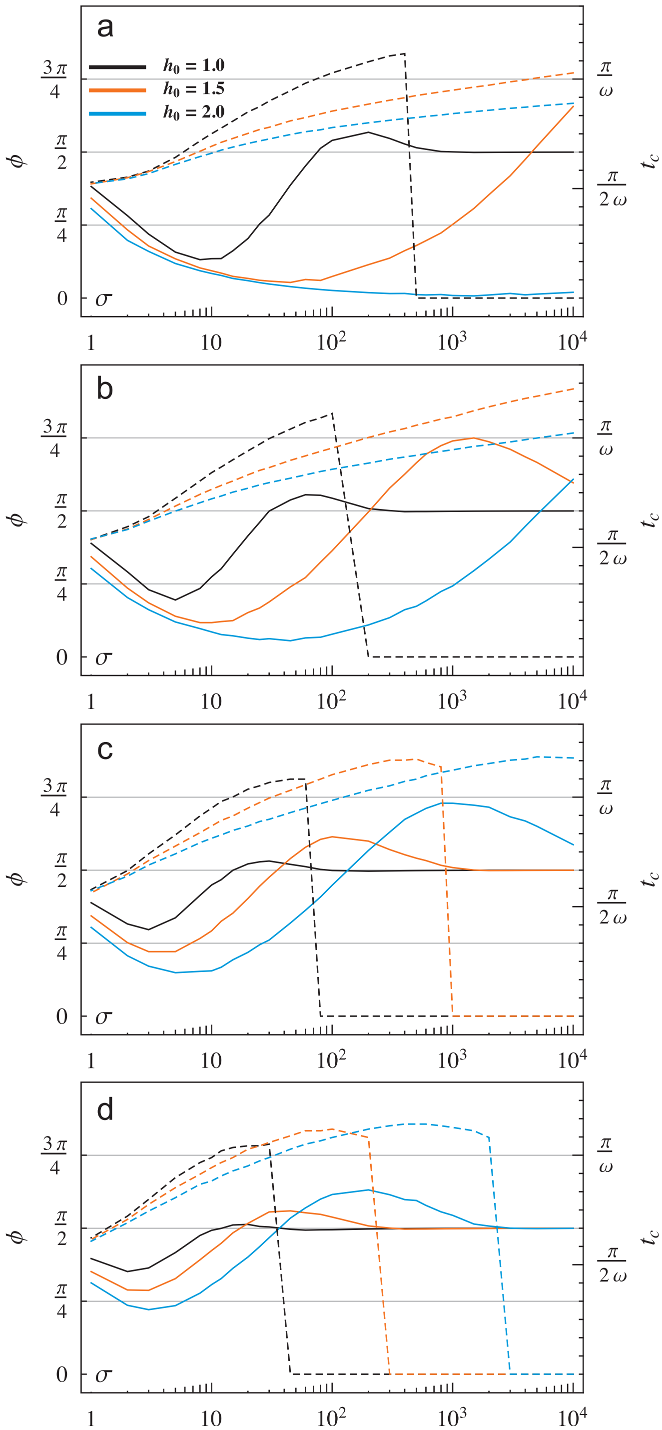

Fig. 2. Phase difference ϕ (full line; left scale) and coercive time t_c (dashed line; right scale) as functions of the inverse temperature parameter σ. Each image shows three curves for $h_0 = 1.0$, 1.5 and 2.0. (a) $\omega = 0.1$, (b) $\omega = 0.2$, (c) $\omega = 0.4$ and (d) $\omega = 0.6$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

hand, increasing the frequency is known to linearize the magnetic response, transforming the loops into quasi-ellipsoids. This can be seen in some of the loops of Fig. 1, usually at low σ. The frequency interval here chosen is midway through this transition, which is usually finished for $\omega \simeq 1$ (not shown) [2].

In order to properly understand the influence of each parameter on the system, we now turn to a quantitative description of the problem, where we study the temperature dependence of the phase lag (ϕ) between the magnetization and the external field, the coercive time (t_c), and also the real and imaginary susceptibilities, defined as

$$\chi' + i\chi'' = \frac{\omega}{\pi h_0} \int_0^{2\pi/\omega} m(t) e^{i\omega t}\, dt. \tag{4}$$

In Fig. 2 we show the results for ϕ (full lines; right scale) and t_c (dashed line; left scale), and in Fig. 3 for χ' (full lines) and χ'' (dashed line; same scale). We begin by describing some general features of these parameters, which are expected to occur for all combinations of ω and h_0. Incidentally, the temperature at which they take place may be too low to have any physical meaning. This is usually the case for low ω and/or high h_0, which is discussed below.

During the superparamagnetic–ferromagnetic transition, ϕ is observed to decrease with σ, until reaching a characteristic minima which we shall refer to as the condition of optimal balance between both competing responses (Fig. 2). It is the situation where thermally induced jumps become minimized while field-assisted jumps remain effective. Consequently, the loop heights approach unity meaning that practically all macrospins are flipping every half-cycle. After this minima the behavior of ϕ becomes very sensitive to ω and h_0. It eventually passes through the value $\phi = \pi/2$ (where the colored dots in Fig. 1 touch the abscissa), corresponding to the temperature at which the loop symmetry is broken. The presence of a maxima before reaching its asymptotic value indicates that this asymmetry is also governed by a competing balance, as previously discussed.

The coercive time equals $\pi/(2\omega)$ at high temperature (zero coercivity) and is seen to increase with σ. As mentioned, it surpasses π/ω at the point where ϕ first reaches $\pi/2$. At sufficiently low temperatures it no longer makes sense to compute a coercive time since the loops have shrank toward the abscissa. Thus, t_c falls abruptly to zero.

The real part of the susceptibility (χ') represents the in-phase component of the magnetization (Fig. 3). Its general features follow very closely those of the phase lag. It shows a maxima near the condition of optimal balance and becomes negative when $\phi = \pi/2$, also showing a minima where ϕ has a maxima. This negativity is a manifestation of the enhanced number of out-of-phase moments and a signature of the dynamic symmetry loss.

The imaginary part of the AC susceptibility (χ'') is observed to be always a single-peaked function of σ (Fig. 3). It is directly proportional to the loop area, thus being reasonable for its maxima not to coincide with that of χ'. For, while the latter is proportional to the loop height, the former depends on both its height and width. The height is maximum near the situation of optimal balance and starts to decrease with increasing σ. But, meanwhile, t_c is increasing so that a point inevitably comes where the area reaches its largest value.

4. Discussion and conclusions

In terms of real measurements one must remember that $\sigma \propto 1/T$. The response of the system, as we have shown, is sensitive to all parameters, but this asymptotic dependence allows for very abrupt changes to occur within very small temperature variations. Consider, for instance, a system in which $\sigma = 1$ corresponds to $T = 300$ K, so that the interval $100 \le \sigma \le 300$ is then equivalent to $3\text{ K} \ge T \ge 1$ K. Taking the example of $\omega = 0.1$ and $h_0 = 1$ (Figs. 2(a) and 3(a), black curves), it can be seen that within just a couple of degrees, the hysteresis loops can go from symmetric to asymmetric. Furthermore, over the same interval, the area of the loops diminish by up to a factor of four and, finally, any conclusions drawn for a given field amplitude have to be completely modified if a different value is used (see, for instance,

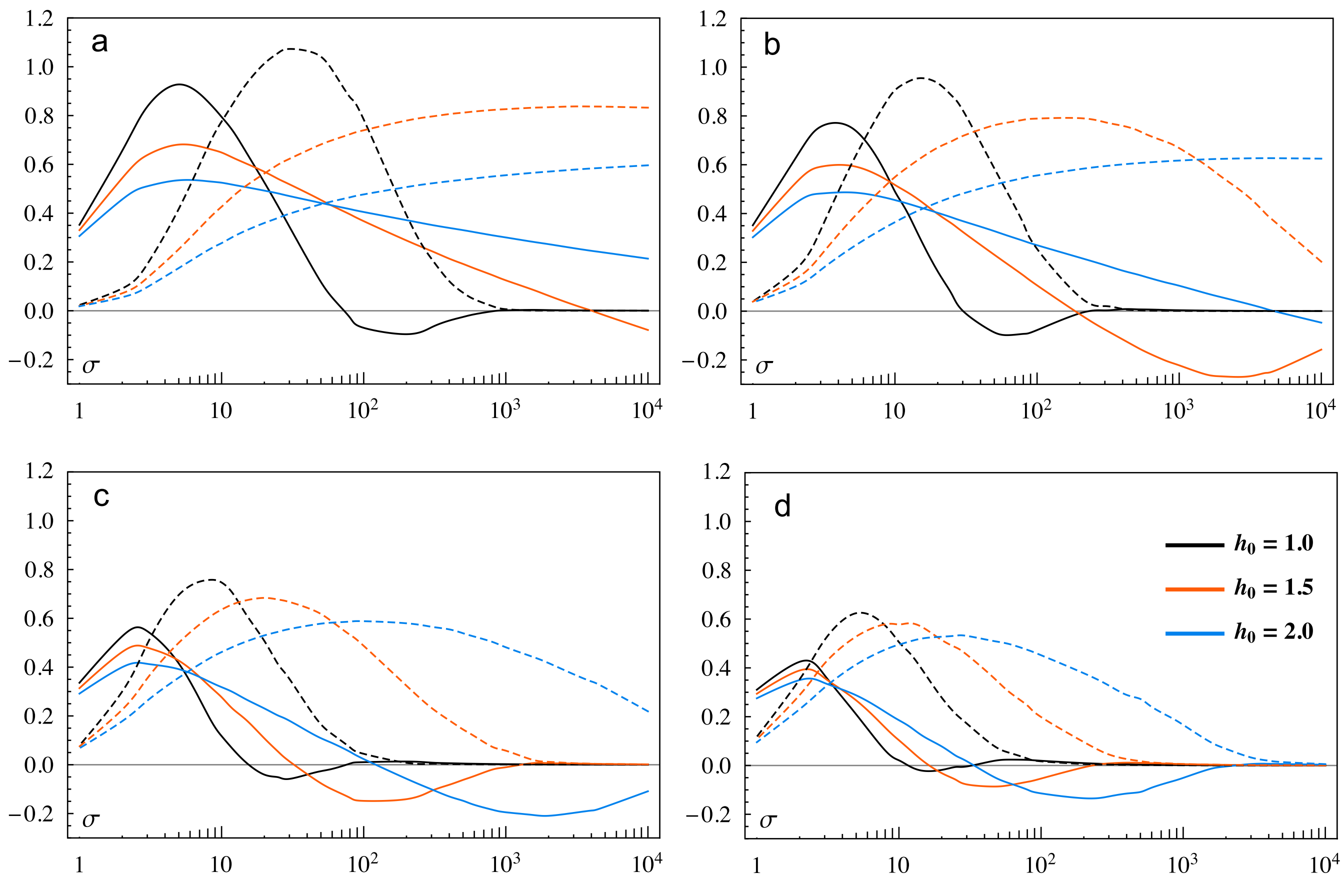

Fig. 3. Real (χ'; full line) and imaginary (χ''; dashed line) parts of the AC susceptibility as a function of σ. Other details are similar to those in Fig. 2. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

$\omega = 0.1$ and $h_0 = 1.5$; red curves). We note in passing that the system can already be considered as thermally stable (ferromagnetic) for $\sigma \simeq 15$, which in our example would mean $T \simeq 20$ K. Notwithstanding, the bulk of interesting phenomena occurs at even lower temperatures.

The values of ω and h_0 employed in these calculations are considerably high. Even so, from the results in Figs. 2 and 3, it can be seen that the superparamagnetic–ferromagnetic phase transition remains predominantly a function of the temperature. In opposition, the dynamic symmetry loss is clearly very sensitive to the characteristics of the magnetic field. At high frequency, such as $\omega = 0.6$ (Figs. 2 and 3(d)), increasing h_0 simply shifts the general properties to lower temperatures while simultaneously enhancing their extrema. At lower frequencies, on the other hand, the symmetry loss may or may not take place in the computed (and physically meaningful) range of temperatures studied. These results strongly suggest that this is indeed a phase transition, akin to that observed in Ising ferromagnets. In view of the different reversal mechanisms governing each system, our results also corroborate the universality of this effect.

Throughout the temperature range studied, a common characteristic of all conditions is the sharp variations in the loop area, which can also be associated with the energy dissipated within each cycle. Its maxima is seen to occur always between the situations of optimal balance and dynamic symmetry loss (Fig. 3). In such, the fractional area defined as $A = \chi''(\pi h_0/4)$ may reach values as high as 80% for $h_0 = 1$ and 90% for $h_0 = 2$. One may thus assert that a third regime also exists, characterized by a large energy dissipation.

In conclusion, we have investigated the dynamic response of single-domain particles upon different combinations of the field amplitude, frequency and temperature. From high to low temperatures, it is possible to observe that the transition from the superparamagnetic to the ferromagnetic states depends weakly on the external field. An optimal temperature is seen to exist, where thermal jumps become suppressed while field-induced jumps remain efficient. Upon further decreasing the temperature one passes through a condition of high energy dissipation (large loop area) and then reaches a point where the magnetic response becomes predominantly out-of-phase. This characterizes a dynamic symmetry loss and results from the thermal fluctuations, which albeit small, enable the magnetic field to maintain a reasonable amount of macrospins flipping during each cycle. In striking opposition to Ising systems, where a similar transition is known to take place, the characteristic sensitivity of single-domain particles to thermal fluctuations allow for this phase-transition to take place over very narrow temperature variations.

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