Dynamic origin of first and second order phase transitions in magnetization reversal of elliptical nanodots

F. Montoncello,[1] L. Giovannini,[1] F. Nizzoli,[1] P. Vavassori,[1,2] and M. Grimsditch[3]
[1]Dipartimento di Fisica, Università di Ferrara and CNISM, Via Saragat 1, I-44100 Ferrara, Italy
[2]CNR-INFM S3, I-41100 Modena, Italy and CIC nanoGUNE Research Center, E-20009 San Sebastian, Spain
[3]Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA
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We study the magnetization reversal in elliptical nanodots with the external field applied exactly along the minor (hard) axis. By varying the magnitude of the applied field, several first and second order transitions take place and the system proceeds through magnetic configurations characterized by different symmetry properties. The dynamical matrix method is used to calculate the spin excitations as function of the applied field. This model system allows us to investigate the relationship between the singularities of the magnetization, the presence of soft spin excitations, and the symmetry properties of the static and dynamic magnetization fields. Rules that govern the transitions are formulated.

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I. INTRODUCTION

The reversal of the magnetization in magnetic films and particles is undoubtedly of great interest for practical applications due to its crucial role in any process for storing or modifying information in magnetic memories and devices. In addition, this subject is very important from the basic point of view. During the process of reorientation fundamental phenomena, such as phase transitions, symmetry changes or singularities in the spin wave spectrum occur. Indeed, the necessary ingredient for observing the phenomenon of reorientation is the presence of some kind of anisotropy (typically, uniaxial or shape anisotropy) that competes with the tendency of the magnetization to align in the direction of the applied magnetic field.

Here, we wish to investigate the relationship between soft mode dynamics and discontinuities in the magnetization. In order to do so, we must distinguish between nonlinear dynamics,[1] which corresponds to large amplitude perturbations, and the linear dynamics of small amplitude magnetic excitations.[2] Here, we refer to the latter case. In this context, our investigation corresponds to the zero temperature case. In the present framework, it is not possible to investigate the entire process of reorientation and the response to an instantaneous magnetic field pulse; our aim is to exploit the relatively simpler case of linear spin dynamics to understand what happens at the onset of the different discontinuities involved in the reversal process.

Although there exists an extensive literature on spin dynamics of magnetic films, the study of small submicrometric particles has only been developed recently.[2] While for infinitely extended films, the in-plane translational invariance permits the development of analytical theories and the derivation of explicit results for both the equilibrium states and the magnetic excitations; a realistic treatment of confined particles requires the inclusion of nonhomogeneous magnetization distributions. Apart from dots of highly symmetrical shape, like cylinders, most of the published studies for confined structures refer to numerical micromagnetic simulations which determine the equilibrium state of the system under the action of a given applied field, and occasionally, the spectrum of the spin modes extracted from the time evolution of the magnetization in condition of very small damping or by introducing a fluctuating (Langevin) field.[2]

In recent years, elliptical dots of different aspect ratios have been considered as interesting systems to study static and dynamic magnetic properties, when the applied field changes in magnitude and/or direction. In particular, the problem of magnetic instabilities and reversal has been tackled both theoretically[3,4] and experimentally.[5,6] Spin excitations in elliptical particles have also been investigated with different methods.[7,8] However no attempt was made to establish a precise connection between magnetic instabilities and soft modes in the spin excitation spectrum.

In a recent paper,[9] we have shown that calculations of the long-wavelength spin excitations provides an excellent description of the spectra of elliptical Permalloy nanodots measured by Brillouin light scattering. We furthermore pointed out the dynamic origin of the reversal process by identifying the spin modes responsible for the onset of the instabilities that lead to reversal. The relevant spin modes soften at the critical fields, i.e., their frequency vanishes, so that also the restoring "forces" vanish and an instability occurs. In order to reproduce the measured hysteresis loops, in Ref. 9, we were obliged to make the assumption that the external field was not exactly applied along a symmetry axis of the ellipse, but was slightly misaligned by $\approx 1°$, mimicking the real experimental situation. The consequence of this assumption is to break the mirror symmetries typical of a perfectly aligned saturated elliptical dot and allow both the static and dynamic magnetic properties of the dots to be quantitatively reproduced. From a theoretical point of view, the disadvantage of a misaligned magnetic field is that the equilibrium configuration maintains the same symmetry at any field (invariance for space inversion) and consequently, strictly speaking, symmetry changes were eliminated from the problem in that study.

In this paper, we consider the applied field to be exactly aligned along the minor (hard) axis of an elliptical dot. In this case, new and nontrivial effects arise because the initial symmetry of the system is high and is broken during reversal. We will show that the magnetization exhibits several discontinuities and symmetry changes. The elliptical dot thus behaves as a model system for investigating the relationship between phase transitions, symmetry changes, and soft mode dynamics in nanometric particles. In Sec. II, the general features of the low frequency spin excitations, calculated vs the applied field, are presented and compared with the behavior of the static magnetization. After a short discussion about the symmetry of the problem, details of how the specific soft modes are correlated with each phase transition in the ground state are given in Sec III. Conclusions are drawn in Sec. IV.

II. NORMAL MODE CALCULATION

We use the dynamical matrix method[10,11] to calculate the magnetic normal modes of an elliptical dot. The method allows us to evaluate both eigenvalues (mode frequencies) and eigenvectors (amplitude distribution) directly, without the need of integrating the Landau–Lifshitz equation and Fourier transforming the time evolution of the magnetization field. The eigenvectors yield the dynamic Cartesian components of the magnetization δm_i (mode profiles).[10] The calculation includes Zeeman, dipole, and exchange terms. We adopt a reference frame with the z axis normal to the particle surfaces, the x axis along the minor axis of the ellipse, and the y axis along the major axis.

We study an elliptical particle with dimensions $200 \times 500 \times 15\ \mathrm{nm}^3$, divided in small cells of size $5 \times 5 \times 15\ \mathrm{nm}^3$, for a total of 3144 cells. The large aspect ratio (2.5) avoids the formation of vortex states.[5] The material parameters used in our simulations are saturation magnetization M_s=800 G, exchange stiffness A=1.3 μerg/cm, and gyromagnetic ratio γ=2.95 GHz/kOe. The external field is applied exactly along the x axis; initially, it points in the positive direction (H=1000 Oe), decreases, and then becomes negative, down to H=−1000 Oe. The calculation of the normal modes requires, for each value of the applied field, the knowledge of the equilibrium state. This has been obtained by using the micromagnetic simulator OOMMF.[12]

The hysteresis half cycle calculated from positive to negative fields is shown in Fig. 1(a) for both the longitudinal (x) and the transverse (y) components of the magnetization. It shows that the total dot magnetization rotates from the positive x axis to the positive y axis, and finally, to the negative x axis. There are three large (at H equal to 530, 397, and −519 Oe) and one small (at H equal to 805 Oe) discontinuities in the magnetization during reversal corresponding to first order transitions. It was shown in Ref. 9 that in a real experiment (and in a calculation with a 1° misalignment of the applied field with respect to the x axis), three of these four discontinuities are smeared out and only the sharp transition at about −500 Oe is still observed. In the previous paper, we showed that the reorientation of the magnetization occurs via a coherent rotation till to about H=−500 Oe. Although some of the modes in Ref. 9 were found to develop frequency minima in the vicinity of rapid changes in M, the frequency of these modes did not reach zero. In the present case, the reorientation toward the easy axis comes about via

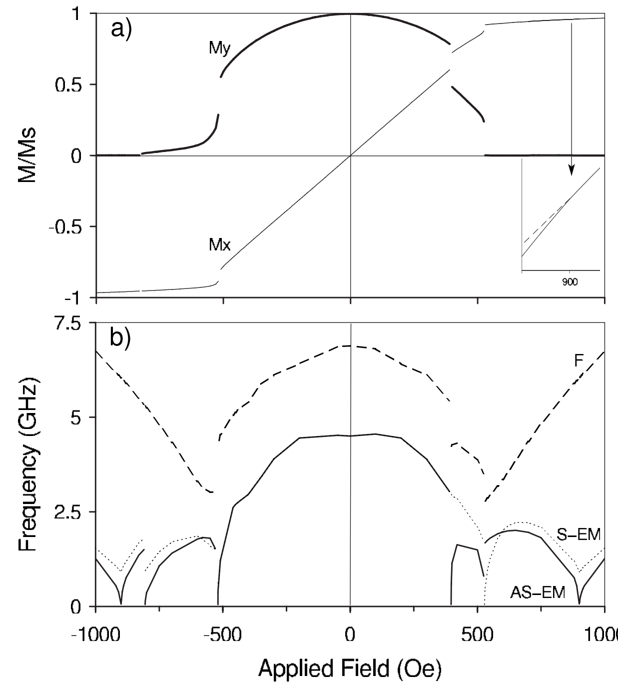

FIG. 1. Upper panel (a): magnetization components M_y (bold line) and M_x (thin line) calculated from positive to negative fields. The inset is a magnification of the behavior of M_x, close to the critical field H_c=901 Oe, exhibiting a kink. The dashed line in the inset is a straight line extrapolating the behavior of M_x for fields greater than 901 Oe. Lower panel (b): frequency of the three spin modes of lowest energy. F: dashed line. S-EM: dotted line. AS-EM: solid line. In the range −519–+397 Oe, the S and AS end modes are almost degenerate, so that only one curve is drawn.

two sudden discontinuities in the range 530−397 Oe. We also find two other transitions (at H equal to 901 and −901 Oe) that are not evident in Fig. 1. We will show that these transitions are also induced by a soft mode but that they are second order transitions and only result in a discontinuity in the first derivative of the magnetization, as shown in the inset in Fig. 1.

In Fig. 1(b), the frequencies of the lowest spin modes (quasiuniform or fundamental mode: F, symmetric end mode: S-EM, and antisymmetric end mode: AS-EM) are plotted as a function of the applied field. Obviously many other modes are provided by the calculation[8,9] but are not shown here because they are of no interest in the present framework. The comparison of the soft modes with the hysteresis cycle clearly shows a correspondence between the discontinuities of the magnetization or of its first derivative and the singularities of the modes. Detailed discussions of the transitions will be given in the following section. We note however, that apart from the soft mode at about −500 Oe, the results with and without misalignment are surprisingly different; these differences can be traced to the very different nature of the equilibrium states that are reached during reversal.

Two other remarks are also needed. First, because hybridization effects are present, there are cases in which certain modes are not “pure,” i.e., when hybridization occurs, the profile of some modes can exhibit features of another mode.

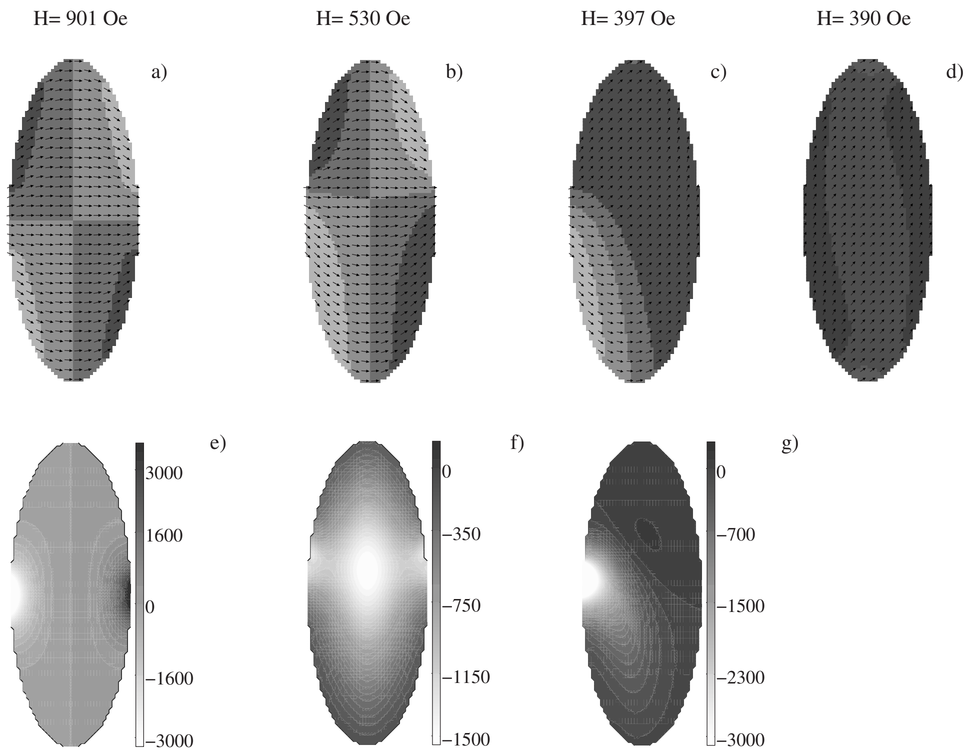

FIG. 2. Upper row: equilibrium states of the static magnetization for a few positive applied fields. H=901, 530, and 397 Oe are critical fields. The gray levels of the background have been chosen to mark the magnitude and sign of the y-magnetization component. (a) Onion state; (b) C state; (c) state with no symmetry elements; (d) S state. Lower row: y component δm_y of the dynamic magnetization for three soft modes calculated for the corresponding equilibrium states.

Second is that the mode profiles that are shown in Figs. 2–4 represent a snapshot of given components of the spin precession. At a later time (e.g., half a precession cycle), the light areas will have become dark and vice versa.

We use S (symmetric) and AS (antisymmetric) to denote the symmetry of modes under inversion operation. Although strictly speaking, this can only be done if the ground state has inversion symmetry, we also use the notation somewhat more loosely to denote modes where the spin motion at opposite ends of the particle is in or out of phase. Notice that the S-EM and the AS-EM in Fig. 1(b) are almost degenerate in the field range $-519-+397$ Oe, while there is an appreciable splitting for larger values of $|H|$ even far from the critical fields. This can be understood by considering that each EM is strongly localized close to opposite edges of the ellipse, along a direction that depends on the value of the applied field.[8] When H is in the range $-519-+397$ Oe, the regions of localization of the end modes are located along the major axis of the ellipse (500 nm) so that they are faraway, do not overlap, and the modes are degenerate. Instead, when H is large and the dot is almost saturated along the x direction, these localized regions located along the minor axis of the ellipse (200 nm) are rather close to each other and do overlap. Therefore, in the latter case, the degeneracy is broken and the splitting is observed.

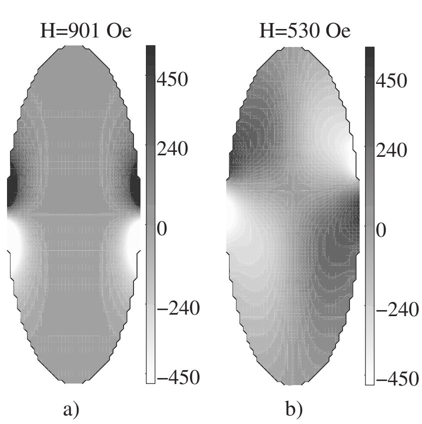

FIG. 3. x component δm_x of the dynamic magnetization for two soft modes calculated at the critical fields H=901 and 530 Oe. Panel (a) corresponds to the y component of Fig. 2(e) and panel (b) corresponds to the y component of Fig. 2(f).

III. PHASE TRANSITIONS

A. Symmetry considerations

Although the ellipse itself possesses D_{2h} symmetry, application of a magnetic field along a principal axis reduces the number of symmetry elements to four, belonging to the point group C_{2h}: besides the identity (E), a twofold rotation axis (C_2) about the field axis, the inversion symmetry (i), and the mirror plane perpendicular to the rotation axis (σ_h). Note that $\sigma_h = i \otimes C_2$. We remind the reader that the pseudovector nature of the magnetization and applied field requires a sign reversal after one of the improper rotations i and σ_h has been applied. For a given ground state, the symmetry of $\boldsymbol{\delta m}$, an eigenvector of a dynamical problem, will belong to one of the allowed representations; for C_{2h}, for example, it will have A_g, B_g, A_u, or B_u symmetry. The latter two are odd under inversion, the B modes are odd under the C_2 operation, and the B_g and A_u are odd under σ_h.[13]

At high fields, where the magnetization aligns parallel to the applied field, even including the remanent onion state, the system has the full C_{2h} symmetry discussed above. As the field is lowered and the magnetization takes on either

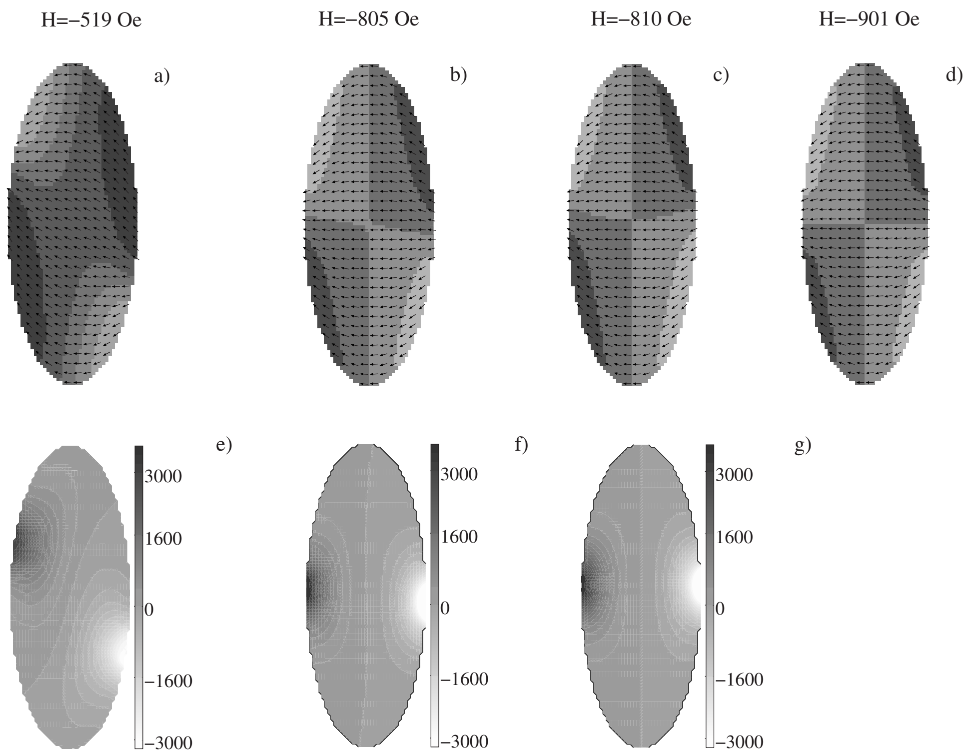

FIG. 4. Upper row: equilibrium states of the static magnetization for a few negative applied fields. The gray levels of the background have been chosen to mark the magnitude and sign of the y component of the magnetization. (a) and (b) S states; (c) C state; (d) onion state. Lower row: y component δm_y of the dynamic magnetization for three modes calculated for the corresponding equilibrium states. $H=-519$ and -805 Oe are critical fields and the corresponding modes are soft modes.

"C" of "S" like character,[2] a few symmetry elements are spontaneously lost and we will show how it is the symmetry of the soft mode that governs this symmetry breaking. Specifically, in a C state, the magnetization retains only the σ_h element and in an S state, only the i element. The reader should be aware that the symbol S, signifying a ground state, should not be confused with the label S which refers to the symmetric character of the modes.

B. Phase transitions

We will discuss the transitions in order as they occur as the field is reduced from a large positive value. From the inspection of the hysteresis cycle [Fig. 1(a)] and of the mode frequency behavior [Fig. 1(b)], the first transition occurs at a critical field $H_c=901$ Oe. Above this value, the "onion" state shown in Fig. 2(a) is the ground state. Below this field, the ground state slowly develops into a C state. By the time the field reaches 530 Oe, the C state is clearly seen in Fig. 2(b). Figure 2(e) shows the profile of the dynamic magnetization (δm_y component) of the AS end mode that goes soft at 901 Oe. The out-of-plane component δm_z is simply proportional to δm_y, apart from a $\pi/2$ phase factor. It is useful to show [Fig. 3(a)] also the other component δm_x because it helps to appreciate the symmetry properties of the mode. One can see that when the restoring force for this mode goes to zero and its displacements [Figs. 2(e) and 3(a)] are added to the ground state [Fig. 2(a)] one of the two possible C states will ensue. In fact, the spontaneous symmetry breaking of the onion state may lead to two degenerate final C states, obtained by the rotation C_2, which is symmetry operation of the initial (onion) configuration. The choice of Fig. 2(b) is simply due to numerical reasons and does not affect the successive discussion.

As mentioned earlier, the ground state shown in Fig. 2(a) has symmetry elements C_2, σ_h, and i. An analysis of Figs. 2(e) and 3(a) shows straight away that the eigenvector $\boldsymbol{\delta m}$ of the AS soft mode is odd under inversion i (according to the B_u representation), while the ground state is even (A_g representation). Direct inspection of the symmetry properties of the ground state and of the soft mode, as well as comparison of the characters of the representation A_g and B_u,[13] show that the common shared symmetry operator is the mirror element σ_h. As a consequence, the new ground state—which must contain all the symmetry operations common to both the original ground state and the soft mode—keeps the σ_h symmetry, i.e., is a C state belonging to the A' representation (even under σ_h) of the group C_s.[13] The state C evolves under the application of the external field and is shown in Fig. 2(b) for $H=530$.

The transition occurring at $H=901$ Oe is of second order, as proved both by the continuity of the soft mode frequency plot, which vanishes on both sides of the critical field and by the behavior of the M_x component of the magnetization of the dot. As can be seen in the inset of Fig. 1(a), for $H=901$ Oe, M_x is continuous but its first derivative is discontinuous. We remark that this second order transition exhibits a behavior very similar to that of spin wave theory for films with in-plane uniaxial anisotropy.[14,15] Uniaxial anisotropy in films plays the same role of the shape anisotropy in our dot. In the case of the film, a discontinuity of the first derivative of the longitudinal component of the magnetization corresponds to a second order transition and a zero gap in the magnon frequency, at least in mean field theory or in the $T\to 0$ limit of the Green's function approach.[15] In our elliptical dots, we have found a similar behavior, i.e., the soft mode frequency vanishes like $\omega_+=A_+(H-H_c)^\alpha$ for $H>H_c$ and $\omega_-=A_-(H_c-H)^\alpha$ for $H<H_c$, with a critical exponent

$\alpha=1/2$ and a ratio A_-/A_+ close to $\sqrt{2}$, as in films.[14,15] We have also found that at the critical field H_c, the axial ratio of the elliptical precession of the soft mode (defined as the ratio $\delta m_z/\delta m_{xy}$ between the out-of-plane dynamic component δm_z and the in-plane component δm_{xy}) goes to zero, in agreement with an instability occurring in the dot plane.

There is another interesting aspect of the transition discussed above. One could wonder what would have happened if the soft mode had been the S-EM instead of the AS-EM. Depending on the size and aspect ratio of the ellipse, this is indeed a possible scenario. If the S-EM goes soft, it is visually clear that an S-like state would ensue. From the symmetry standpoint, we note that the S-EM is even with respect to inversion i; following the arguments given above, we would expect the new ground state to lose the σ_h mirror symmetry but retain the inversion symmetry. Indeed, the resulting S state does have inversion symmetry. An example of an S state is shown in Fig. 4(b).

It is worthwhile to note that in the case of a small misalignment of the applied field, as in the calculations in Ref. 9, this second order phase transition disappears because the full symmetry of the onion ground state is broken for any applied field and the equilibrium state is already a rotated S-like state even for strong applied fields.

The next transition in the hysteresis loop is at 530 Oe and is again driven by a soft mode, which we loosely call a S-EM. In this case, the behavior is typical of first order transitions. We have numerically verified that, before the critical field, the frequency of the soft mode goes to zero with the same square root dependence already found for the second order transitions. Also, the ratio $\delta m_z/\delta m_{xy}$ vanishes. The nature of the transition can be understood by inspection of Fig. 2. Figure 2(b) shows the ground state just prior to the discontinuity. The profile of the soft mode, shown in Figs. 2(f) and 3(b) for δm_y and δm_x, respectively, shows elements of hybridization of the uniform and a S-EM. The eigenvector $\boldsymbol{\delta m}$ belongs to the A'' representation (odd under σ_h) of the group C_s,[13] and since the ground state belongs to the A' representation, the combination of a ground state and a soft mode with symmetry elements of void intersection leads to a ground state with no symmetry. Equivalently, we can also state that the soft mode of magnetization $\boldsymbol{\delta m}$ does not possess symmetry operations because the symmetry operation of the ground state does not bring $\boldsymbol{\delta m}$ back to itself. The new ground state evolves into the state shown just above the next transition at 397 Oe in Fig. 2(c). The soft mode at this field is shown in Fig. 2(g) and it induces a transformation to the state shown in Fig. 2(d), which is a rotated S-like state with inversion symmetry i, belonging to the even A_g representation of the group C_i.[13] This state is identical to the state that evolved continuously from the high fields when the field was misaligned by 1°.[9] It is worth emphasizing that that simple misalignment eliminated all three of the phase transitions discussed up to now. The latter transition is an example of "symmetry raising," with a final ground state with more symmetry elements than the initial state. This process is reached by a local reorientation of the magnetization in the lower-left quadrant of the ellipse, evidenced by comparing Fig. 2(c) and 2(d).

As already shown in Ref. 9, between 397 and –519 Oe, the system evolves by a coherent rotation of the S-like state, shown in Fig. 2(d). The S-like ground state at $H=-519$ Oe is shown in Fig. 4(a). As already explained in the previous section, in the range from 397 to -519 Oe, the S and AS end modes are nearly degenerate in frequency, as it is visible in Fig. 1(b), where only one end mode is drawn for clarity.

The next first order transition occurs for $H=-519$ Oe, as shown in Fig. 1. The ground state and the soft mode profile at $H=-519$ Oe are shown in Figs. 4(a) and 4(e), respectively. The ground state is invariant for space inversion (it is an S state), while the soft mode has no symmetry compatible with the ground state. As argued above, the resulting new ground state has no symmetry. However, on decreasing the applied field by a few oersteds, this new state again becomes unstable. We have found that this instability is also driven by a soft mode that, for the sake of simplicity, will not be discussed here. The resulting ground state is again an S state, of the kind shown in Fig. 4(b).

By further decreasing the applied field (viz. making it more negative), the new S state becomes unstable at -805 Oe, giving rise to yet another first order transition, as witnessed by the small discontinuity of the y component of the magnetization [Fig. 1(a)] and a corresponding soft mode [Fig. 1(b)]. Just before the transition, for $H=-805$ Oe, the calculated magnetization of the ground state is shown in Fig. 4(b) and the profile of the soft AS-EM in Fig. 4(f). The S ground state possesses inversion symmetry i, differently from the AS-EM which has no compatible symmetry. Since the intersection is void, the symmetry operation i is lost. This actually happens, but the novelty is that the new ground state [Fig. 4(c)] has acquired the symmetry σ_h. It is therefore a C state, belonging to the A' representation of C_s, already met in a previously discussed transition.

By increasing the modulus of the applied field, the ground state continuously develops into the onion state, which is finally reached for $H=-910$ Oe [Fig. 4(d)]. This transition is second order because it is driven by a soft mode [Fig. 4(g)] that goes continuously to zero frequency on both sides of the transition. It is the reciprocal of the transition at $H=901$ Oe, so that it does not deserve explicit discussion.

The outcome of our calculations allows us to draw important conclusions concerning the symmetry of the soft modes and the evolution of the system at the onset of the transitions they drive. If at a critical field, one mode becomes soft so that its symmetry controls the transition, the new ground state after the transition will contain all the symmetry operators common to both the original ground state and the soft mode. If the intersection of the symmetry operators is void, the final state either has no symmetry or it gains a symmetry not present in the original ground state before the transition (symmetry raising). In the latter case, it is not possible to make *a priori* a prediction on the basis of the soft mode only and the final ground state depends on the nearest available states in the phase space.

IV. CONCLUSIONS

An elliptical Permalloy dot subjected to an external field applied *exactly* along the minor axis of the ellipse proved to be an excellent model system to investigate a significant number of phase transitions between magnetic states differing from each other by the intrinsic symmetry properties of the magnetization and to draw important rules on symmetry. Since the symmetry operations we consider here include improper rotations such as space inversion and reflection at a mirror plane, it is necessary to recall that the magnetization vector is an axial vector and must be treated accordingly. The frequencies and the profiles of the spin excitations are calculated in the framework of micromagnetics by the dynamical matrix method, i.e., directly in the frequency domain. We have found the existence of a one-to-one correspondence between magnetic phase transitions and soft modes, i.e., spin excitations of vanishing frequency. This demonstrates the intimate relationship between the dynamics of small amplitude excitations of the nonhomogeneous magnetization in confined structures and the onset of phase transitions. In the sense that we have dealt only with small amplitude spin waves, our approach should be seen as a zero temperature approximation. At finite temperatures, where the amplitudes of the soft modes could be large, nonlinear effects in the dynamics of the spin modes themselves could also affect the reversal mechanisms.

The model system turns out to be particularly interesting because it shows both first and second order transitions. In the former case, the frequency of the soft mode is discontinuous and vanishes only on one side of the transition. For the second order case, it approaches zero continuously on both sides of the transition. On the basis of physical arguments, relating the symmetry properties of the equilibrium states and the corresponding excited states that are corroborated by the analysis of the numerical simulations, we have formulated rules concerning the symmetry changes across each critical field, as a function of the symmetry properties of the initial state and of the soft mode. These considerations hold when the system reverses via equilibrium states having different symmetries, such as the onion, C, and S states of the elliptical dot studied here. Consequently, this can happen only for highly symmetric systems, e.g., in our case, an object with elliptical symmetry with the external field aligned exactly along with one of the principal axes of the ellipse. When the states accessible to the system are all of the same symmetry (e.g., for an ellipse in a misaligned applied field), phase transitions clearly lead to no symmetry changes. Nevertheless, first order phase transitions may also occur, triggered by a soft mode, whenever an abrupt rotation of the magnetization within a region of a dot produces a discontinuity in the total magnetization.

The present kind of investigation provides a deeper understanding of the outcome of more realistic calculations attempting to simulate actual experimental situations.

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