Controlling Double Vortex States in Low-Dimensional Dipolar Systems

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The reversal process of the chirality of each opposite vortex belonging to a double vortex state in ferromagnetic hysterons, via the application of in-plane magnetic fields, is reported. Simulations reveal that such a process involves the formation of four intermediate states, including original ones. Hysteresis loops can occur only in a counterclockwise fashion because of one of these intermediate states. Double vortex states can also be controlled by electric fields in *ferroelectric nanostructures* of different shapes, but with some key differences with respect to the ferromagnetic case.

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Low-dimensional ferromagnets and ferroelectrics are receiving a lot of attention because of their technological promise to yield devices operating at the meso- or nanoscale [1–5]. For instance, the ability to reverse their magnetization or polarization offers the possibility to use the up and down directions of these vectors in nanomemory applications [3,6,7]. However, the magnetized or polarized states in low-dimensional dipolar systems [8,9] can disappear in favor of single vortex states characterized by a finite toroidal moment [8,10]. Recent activity has focused on the possibility of controlling the chirality of such vortices [1,8–12] to design devices of unprecedented capacities [9,13]. Unfortunately, such exciting perspectives do not resist the formidable dependency of dipolar systems' properties on their morphology and size. As a matter of fact, some magnets and ferroelectrics do not exhibit either a polarized structure or a single vortex state, but rather possess a *double* vortex state for which the coexisting two vortices have opposite chiralities [1,9,14,15]. The magnetization or polarization *and* toroidal moment vanish in this latter peculiar state, which is rather associated with a new order parameter, the hypertoroidal moment [16]. This multipole is different in symmetry from the toroidal moment, implying that the fields that are able to control toroidal moments *cannot* control hypertoroidal moments. One may therefore wonder if and how double vortices can be switched in dipolar systems. The aims of this Letter are (i) to prove that such control can be accomplished by straightforward fields, and (ii) to reveal atomistic features of previously unknown intermediate states that allow such control in a definite way.

Let us first focus on the ferromagnetic systems schematized in Fig. 1(a). They consist of two identical disks of radius R merged together, with w and L representing the thickness of the system and half of the distance between the centers of the two disks, respectively (L and w define the y and z axis, respectively). Such low-dimensional structures are called hysterons [15,17], which have been fabricated in Ref. [15] and can exhibit a double vortex state [15]. To study them, we use the hybrid approach of Ref. [12] that combines both atomistic and continuum features. The investigated ferromagnet is thus divided into equal regions containing several unit cells. The total magnetic moment of any of such region j, μ_j, is equal to the sum of the $\mathbf{m}_{i,j}$ local magnetic moments of the magnetic atoms i belonging to that region, assuming that the $\mathbf{m}_{i,j}$ are all identical inside a given region. The total energy of the studied zero-dimensional ferromagnetic structures under an ac magnetic field, $\mu_0\mathbf{H}$ (with μ_0 being the permeability of vacuum), is given by

$$E_{\mathrm{magn}} = 1/2\sum_{jk\alpha\beta} D_{j\alpha,k\beta}\mu_{j\alpha}\mu_{k\beta} - \mu_0\mathbf{H}\cdot\sum_j \boldsymbol{\mu}_j + 1/2J'\sum_{jk\alpha}\mu_{j\alpha}\mu_{k\alpha},$$

where the sums run over the cells j and k and over the Cartesian components α and β. $D_{j\alpha,k\beta}$ is the tensor asso-

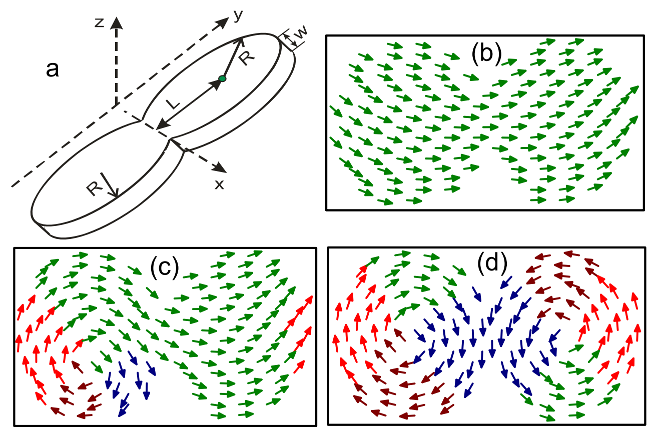

FIG. 1 (color online). Schematization of the investigated ferromagnetic system (a) and its ground-state in a (x, y) plane for different characteristic sizes (b)–(d). Panel (b) corresponds to $R \simeq 392$ nm, $L \simeq 343$ nm, and $w \simeq 98$ nm, while such sizes have all been increased by a ratio of 1.4 and 2.38 in panels (c) and (d), respectively.

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ciated with the long-range magnetic dipole-dipole interactions [12], and the sum over k in the last term only runs over the first nearest neighbors of the cells j. Our ferromagnetic systems are mimicked to be made of Permalloy 80 (i.e., $Ni_{80}Fe_{20}$) by using the relevant exchange parameters [12,18]. E_{magn} is then used to solve the Landau-Lifshitz molecular dynamics equations [19] for all the $\boldsymbol{\mu}_j$ (our method technically differs from the approaches previously used for low-dimensional ferromagnets [20–22]).

For relatively small sizes and when no magnetic field is applied, the ground state of the investigated hysteron is magnetized along the long axis [see Fig. 1(b)]. When increasing the characteristic sizes, the dipole pattern evolves into the peculiar state that has been discovered in Refs. [15,17], and that exhibits a dipole vortex in only one of the two disks while the other disk still possesses a magnetized configuration [see Fig. 1(c)]. Finally, when further increasing sizes, the double vortex state appears, as displayed in Fig. 1(d), for $R \simeq 1349$ nm, $L \simeq 1178$ nm, and $w \simeq 333$ nm. Such a double vortex state has been observed in ferromagnetic hysterons made of permalloys [15] and in other low-dimensional magnets [1,14].

Let us now reveal the response of a double vortex to an ac magnetic field applied along the (short) x axis. For that, in addition to calculate the magnetization $\mathbf{M}$, it is useful to compute the hypertoroidal moment $\mathbf{h}$. Such moment is the sole order parameter of double vortex structures, and also quantifies the curvature of dipole patterns in complex states [16]. It takes the following expression [16]:

$$\mathbf{h} = 1/\{4N\upsilon\}\sum_i\Big\{[\mathbf{r}_i \times (\mathbf{r}_i \times \delta\mathbf{d}_i)] - 1/N\sum_j[\mathbf{r}_i \times (\mathbf{r}_j \times \delta\mathbf{d}_j)] - 1/N\sum_j[\mathbf{r}_j \times (\mathbf{r}_i \times \delta\mathbf{d}_j)]\Big\},$$

where the sums run over the cells, N is the number of these cells, υ is their volume, and $\mathbf{r}_i$ locates their centers, while $\delta\mathbf{d}_i$ is the *difference* between the dipole moment at cell i and the averaged dipole moment. Figure 2 shows the (sole) x-Cartesian components of the magnetization M_x and magnetic hypertoroidal moment h_x as a function of H_x/H_m, at a temperature of $\simeq$100 K for the system adopting the ground state of Fig. 1(d). $\mu_0 H_x$ is the magnetic field, and sinusoidally varies in time between $-\mu_0 H_m$ and $+\mu_0 H_m$ (with $\mu_0 H_m = 40$ mT while the field frequency is 0.15 MHz [23]). Figure 3 provides snapshots of ten important states occurring during these loops. State (1) happens when $H_x = 0$, and it is the double vortex state of Fig. 1(d). It is therefore associated with a vanishing magnetization and a strongly *negative* h_x (because the vortex centered on the right disk is rotating counterclockwise while the vortex centered on the left disk is rotating clockwise). When $\mu_0 H_x$ slightly increases, the centers of the two vortices move towards each other along the y axis to create a magnetization along the x axis and to reduce the magnitude of h_x via the enlargement of the "up" domains located at the extreme two sides of the hysteron. When these two centers become separated by a critical distance of about $2(R - L)$, they further move along *different* directions: one center now moves towards the V-shaped junction of the two disks, while the other vortex center moves towards the Λ-shaped junction, to prevent the system from possessing too many dipoles having opposite directions within a small distance. Such a configuration forms state (2), which has never been previously mentioned and is named here the "tilde" state because of the resemblance between the shape formed by some lines of dipoles and the tilde character. (Note that it would be interesting to use the fractional vortex model [24] to characterize the topology of this new state.) When further increasing $\mu_0 H_x$, vortices abruptly disappear in favor of state (α), which can be classified as a double onion state since each disk exhibits a configuration that bears resemblance with a (single) onion state [1,9,25] (we are not aware of any previous discovery of double onion states). When further increasing $\mu_0 H_x$ up to $+\mu_0 H_m$, state (α) continuously evolves into state (3), by making all the dipoles lie closer to the x axis. This results in a large M_x and vanishing h_x. Interestingly,

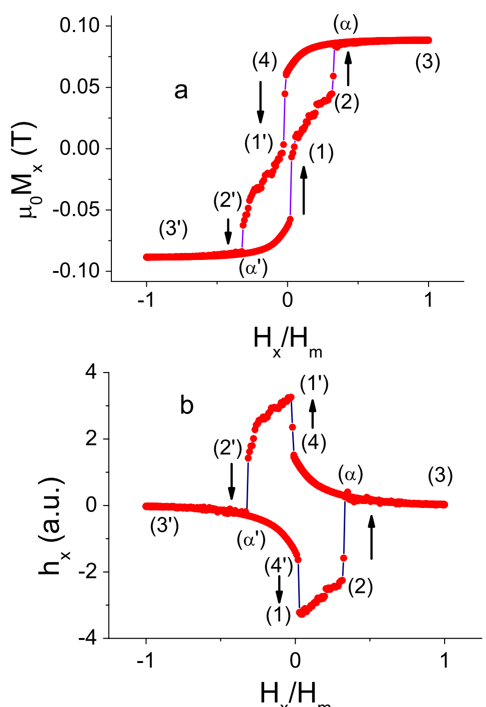

FIG. 2 (color online). Predicted hysteresis loops in the studied ferromagnetic hysteron at 100 K. Panel (a) displays the behavior of the magnetization, while Panel (b) shows the hypertoroidal moment, as a function of the applied homogeneous ac magnetic field.

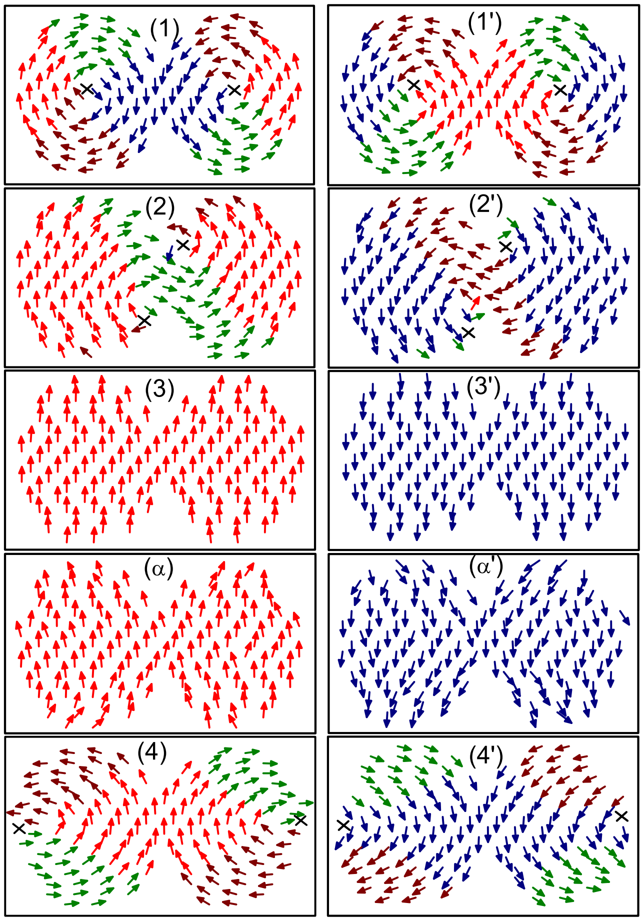

FIG. 3 (color online). Schematization of the dipole arrangement in a (x, y) plane for magnetic states playing a key role in the reversal of the hypertoroidal moment. Crosses represent the vortex centers.

decreasing the field after it reaches its maximum positive value makes state (3) return to state (α) and then leads to a new configuration, that is, state (4). Such latter configuration arises from the simultaneous nucleation of a counterclockwise vortex centered at the extreme left side of the left disk and of a clockwise vortex centered at the extreme right side of the right disk. (Note that the fractional vortex model [24] will also be interesting to use to further describe this nucleation.) h_x is large and *positive* in state (4) because state (α) acts as a seeding state for state (4) and because state (α) has a positive hypertoroidal moment resulting from its specific dipole pattern's curvature. We name the previously unknown state (4) as the χ state. When further decreasing the magnitude of the field but still keeping the positive sign for its x component, the centers of the two vortices of state (4) move toward each other along the y axis, until the double vortex state ($1'$) forms when the field vanishes. State ($1'$) differs from state (1) by exhibiting a vortex of *opposite* chirality in each of the two disks, as consistent with the strong positive hypertoroidal moment in state ($1'$) and its null magnetization. The evolution of state ($1'$) to states ($2'$), (α'), ($3'$), (α'), ($4'$), and then back to state (1), when the field first becomes negative in sign, then reaches its lowest possible negative value, and finally increases towards zero, is similar to the evolution from state (1) to state ($1'$), via states (2), (α), (3), and (4), described above for the positive fields—since state (i$'$) is deduced from state (i), for $i = 1, 2, \alpha, 3$, or 4, by a mirror symmetry about the (y, z) plane passing through the center of gravity of the hysteron. Figures 2 and 3 thus reveal that the chirality of each opposite vortex forming the double vortex state can be controlled by applying an homogenous magnetic field parallel to the x axis, via the formation of the tilde, double onion, homogeneous, and χ states. Interestingly, we further found that starting from state (1) and first applying a negative field results in the path (1)-($4'$)-(α')-($3'$)-(α'), and then back to state (1), before the hysteresis loops of Fig. 2 occurs again. In other words, one cannot go from state ($3'$) to state ($2'$) under a negative homogeneous field. This is because the system energetically prefers to exhibit the double onion state (α') that is associated with a negative h_x as its ground state, for a negative magnetic field of large enough magnitude to result in a large negative magnetization but not too large to also

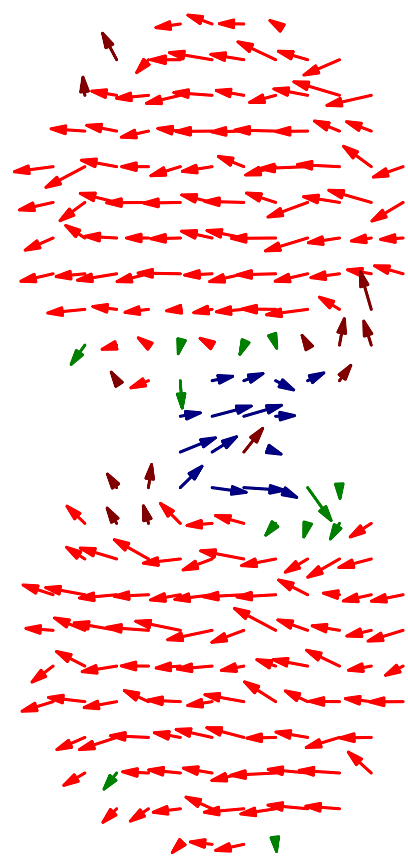

FIG. 4 (color online). The key intermediate state in ferroelectric hysterons. Such a state is the equilibrium phase for large enough electric field applied along the x axis, but not too large either to exhibit a nonvanishing hypertoroidal moment.

have a nonvanishing hypertoroidal moment. The hysteresis loops of Fig. 2 can thus only occur in a counterclockwise fashion, and the control of the hypertoroidal moment's sign is inherently linked to the curvature of the dipoles in states (α) and (α') [26].

Let us now briefly turn our attention to a nanoscale *ferroelectric* hysteron made of $Pb(Zr_{0.4}Ti_{0.6})O_3$, under open circuit electric boundary conditions, as simulated by the effective Hamiltonian approach successfully used in Refs. [12,27]. We chose $R = 64$ Å, $L = 12$ Å, and $w = 24$ Å, because such small sizes already result in a ground state that is a double vortex. An ac electric field applied along the x axis is also found to be able to control the (electric) hypertoroidal moment, but with one major difference with respect to the magnetic case: the only possible way for the hysteresis loops of the polarization-versus-electric field and electric hypertoroidal moment-versus-electric field to occur is in a *clockwise* way. This is because the ferroelectric state corresponding to the magnetic state (α) has a negative hypertoroidal moment. Such a ferroelectric state is shown in Fig. 4, and it possesses a small region centered at the junction of the two disks that exhibits dipoles lying in the opposite direction with respect to its polarization and can therefore be denoted as a ferroelectric bubble state [28]. It is the specific rotation of the dipoles around this small region that yields the negative sign of the hypertoroidal moment [29]. The difference in morphology between such a bubble state and magnetic state (α) originates from the low cost of short-range interactions with respect to depolarizing effects in ferroelectrics, unlike in magnets. We further found that ferroelectric dots elongated along the y direction also possess a double vortex ground-state structure [16] and a bubble state, when applying an ac electric field oriented along an axis perpendicular to the y direction, in the plane of the vortices—which also allows the control of the hypertoroidal moment. In other words, homogeneous fields are capable of controlling double vortices in magnetic and ferroelectric systems *of different shapes*, with such control occurring thanks to specific dipole pattern features (characterized by the hypertoroidal moment) of original intermediate states. These findings may open the way for new technologies in memory devices of nanoscale and mesocale dimensions.

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