Geometric signature of reversal modes in ferromagnetic nanowires

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Abstract – Magnetic nanowires are a good platform to study fundamental processes in magnetism and have many attractive applications in recording such as perpendicular storage and in spintronics such as non-volatile magnetic memory devices (MRAM) and magnetic logic devices. In this work, nanowires are used to study magnetization reversal processes through a novel geometric approach. Reversal modes imprint a definite signature on a parametric curve representing the locus of the critical switching field. We show how the different modes affect the geometry of this curve depending on the nature of the anisotropy (uniaxial or cubic anisotropy), demagnetization and exchange effects. The samples we use are electrochemically grown nickel and cobalt nanowires.

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Introduction. – A surge of renewed interest in ferromagnetic nanowires has occurred very recently triggered by their interesting properties with respect to spintronic devices and more specifically to non-volatile memory (MRAM) and logic devices [1–3].

From a fundamental point of view, they represent a quasi-one-dimensional system displaying magnetic properties in sharp contrast to the Mermin-Wagner [4] theorem forbidding (Heisenberg-type) magnetism in systems of dimension \((D \leq 2)\) with short-range interactions.

They are simpler than nanotubes since their physical properties do not depend on chirality and they can be grown in a variety of methods: molecular beam epitaxy, electrochemical methods (template synthesis, anodic alumina filters), chemical solution techniques (self-assembly, sol-gel, emulsions, etc.) etc.

Template synthesis is an electrochemical method used in the manufacturing of nanostructured materials, in particular, nanowires can be obtained by filling a porous polycarbonate membrane, which contains a large number of cylindrical holes that are track-etched and possess a narrow size distribution.

Individual nanowires as well as arrays of nanowires are important. Characterization and understanding of the magnetic properties of nanowire arrays are challenging since we have dipolar interactions between wires in addition to anisotropy, magnetostatic and exchange effects within individual wires.

Many questions remain open regarding the detailed mechanisms responsible for the magnetization reversal. The intrinsic properties of nanowire arrays are directly related to the properties of the nanoporous template such as the relative pore orientations in the assembly, pore size and its distribution, as well as interpore distance.

Two reversal modes have been suggested as being important: curling and coherent rotation depending on the value of the radius \(a\) with respect to a cutoff value also called “coherence” radius \(R_{coh}\).

For wire radius \(a > R_{coh}\), the reversal occurs by curling. For \(a < R_{coh}\), coherent rotation is expected as predicted by the Stoner-Wohlfarth model [5].

The remanent magnetization distribution within a nanowire depends on its radius as well as on the balance between different contributions (anisotropy, magnetostatic/shape and exchange) to the magnetic energy. Exchange energy dominates at small wire radius and favors a uniform magnetization distribution or non-uniform flower, S, Landau, leaf, etc., states. On the other hand, this almost collinear spin alignment leads to large demagnetizing fields due to magnetic surface charges and correspondingly increase of the magnetostatic energy which increases gradually with radius. When the radius increases beyond the single domain limit \((a > R_{sd})\), the competition between these two energies and anisotropy energy leads to non-uniform magnetic states such as

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multi-domain states (if the anisotropy constant is greater than the magnetostatic energy). It might lead to magnetic vortices, if the anisotropy energy is small in comparison with the magnetostatic energy.

In this work, we set out to study the geometric signature of reversal modes in nanowires depending on the different magnetic properties inherent to the wire belonging to any of the anisotropy, magnetostatic and exchange classes. This paper is organised as follows: in the second section, the geometric signature is discussed in the case of an infinite cylinder with a uniaxial anisotropy. In the third section, the resulting parametric curve is considered for a general ellipsoid with arbitrary uniaxial anisotropy and finally the fourth section bears our conclusions.

Geometric signature of an infinite cylinder. — The geometric signature of a reversal mode is a parametric curve derived from the nanowire magnetization configuration that can be determined from the Brown equation by minimizing the total free energy [6].

In bulk ferromagnetic materials, the energy of the system can be minimized by forming multiple magnetic domains within which the magnetic moments are aligned. However, there is a critical size \( R_{sd} \) below which a particle remains in a single-domain state during switching; it is given by the implicit form: \( R_{sd} = \left( \frac{A}{2\pi N_s M_s} \ln(\frac{2R_{sd}}{a_0}) \right)^{1/2} \) derived by Frei et al. [7] after approximating the nanowire by an ellipsoid of revolution with major axis \( c \) and minor axis, \( a = b \neq c \) and comparing the exchange energy averaged over the ellipsoid volume to the average magnetostatic energy (see fig. 1). For nickel, the following parameters might be used: \( M_s = 485 \text{ emu/cm}^3, a_0 = 0.249 \text{ nm} \) [8].

The curling reversal mode is the dominant magnetization reversal process in magnetic nanowires. The magnetization curling mode was defined by Frei et al. [7] and after it has used for different structure to investigate the magnetic switching of films, spherical particles, prolate ellipsoid and cylinders [9].

The coherence radius \( R_{coh} \) separating uniform rotation and curling is given by: \( R_{coh} = \frac{A}{\pi^2} \sqrt{\frac{2}{\pi a_0}} \), where \( A \) is the exchange stiffness \( (A = 1.5 \times 10^{-6} \text{ erg/cm} \text{ for bulk nickel and cobalt}) \), \( M_s \) is the saturation magnetization and \( N_s \) is the demagnetization coefficient along the nanowire radius (see fig. 1). The parameter \( q \), is the smallest positive zero of the first-kind Bessel function \( J_1(x) \) derivative \( \frac{dJ_1(x)}{dx} \) [10] \( \text{i.e. the smallest positive maximum} \) and the smallest of the derivatives of all ellipsoidal harmonics. It has been evaluated by Aharoni [11] to a 10^{-7} accuracy. He also found an accurate interpolation formula for any aspect ratio \( m = c/a : q = \sum_{i=0}^{n} \frac{b_i}{m} \) with coefficients: \( b_0 = 1.84120, b_1 = 0.48694, b_2 = -0.11381, b_3 = -0.50149, b_4 = 0.54072, b_5 = -0.17200 \), that are accurate to 10^{-6}.

For particle size larger than \( R_{coh} \), yet smaller than \( R_{sd} \), magnetization reversal proceeds through curling. In this mode, magnetization switching is an abrupt process, and the switching field is very close to the nucleation field for all angles.

In the case of an infinite cylinder with a uniaxial anisotropy along the axis \( K \), two distinct calculations were made independently by Chung and Muller [12] and Ishii [13] 20 years apart.

The curling equations to be solved are

\[
\frac{H}{2\pi M_s} \sin(\theta - \phi) = - \left[ \frac{1}{4} + \frac{K}{4\pi M_s^2} \right] \sin 2\theta,
\]

\[
\frac{H}{2\pi M_s} \cos(\theta - \phi) = \frac{1}{2} \sin^2 \theta - \alpha - \frac{K}{4\pi M_s^2} \left[ \frac{\cos 2\theta + \cos^2 \theta}{1 + \cos^2 \theta} \right].
\]

The constant \( \tilde{k} = \frac{7 \times 72}{11 \times 21 \times 38} \) that appears in \( \alpha = \tilde{k} \frac{A}{4\pi} \) is a rational approximation to the first zero of the Bessel function derivative \( \frac{dJ_1(x)}{dx} \) as found by Ishii [13]. The parameter \( S \) is the reduced radius defined as \( S = a/\ell_{ex} \), with \( \ell_{ex} \) the exchange length\(^1\) considered as an intrinsic length scale of the nanowire (e.g., \( \ell_{ex} = 20.6 \text{ nm for nickel} \)). \( S = 0 \) in coherent rotation (Stoner-Wohlfarth [5] limit), \( S = 1 \) in buckling, whereas \( S > 1 \) in curling with the corresponding parametric curves displayed in fig. 2.

\(^1\)The exchange length \( \ell_{ex} = \sqrt{A/K} \) with \( A \), the exchange stiffness constant \( (A \sim 10^{-4} \text{ erg/cm}) \) is obtained from the comparison between the exchange energy density \( (\text{erg/cm}^3) \) \( \frac{A}{4\pi M_s^2} \) and the anisotropy energy density \( \frac{K}{M_s^2} \) with \( K \) representing the (second-order) anisotropy constant of the material. In soft materials whose anisotropy constant \( K \sim 0 \), one uses the magnetostatic exchange length defined as \( \ell_{ex} = \sqrt{A/2\pi M_s^2} \) since \( K \sim 0 \).
The switching field components $h_x = \frac{H \sin(\phi)}{2\pi M_a}$, $h_z = \frac{H \cos(\phi)}{2\pi M_a}$ are then obtained as

$$
\begin{align*}
    h_x &= \sin \theta \left[ 1 - \alpha + \frac{K \sin^2 \theta}{\pi M_a^2 (1 + \cos^2 \theta)} \right], \\
    h_z &= -\cos \theta \left[ \alpha - \frac{2K \sin^2 \theta}{\pi M_a^2 (1 + \cos^2 \theta)} \right].
\end{align*}
$$

The above equations do not produce the coherent rotation limit as $S \to 0$ in which case, one writes

$$
\begin{align*}
    h_x &= \left( \frac{K}{2\pi M_a^2} + 1 \right) \sin^3 \theta, \\
    h_z &= -\left( \frac{K}{2\pi M_a^2} + 1 \right) \cos^3 \theta.
\end{align*}
$$

There is a discrepancy in the results obtained by Ishii [13] and Chung and Muller [12] whose $\alpha$ and anisotropy $K$ factors are off by a factor of $\frac{1}{2}$.

Geometric signature of a finite ellipsoid with arbitrary uniaxial anisotropy. – Let us consider the general case of an ellipsoid with fourth-order uniaxial anisotropy characterized by two constants $K_1$ and $K_2$. Following Aharoni [9], Wegrowe et al. [14] performed a detailed study of template synthesised nickel nanowires and concluded that the anisotropy of their nanowires is positive and large (recall that for bulk nickel, the lowest-order cubic anisotropy constants are $K_1 = -4.5 \times 10^4$ erg/cm$^3$ and $K_2 = 2.3 \times 10^4$ erg/cm$^3$ at room temperature) arguing that a large strain acting on the nanowires may induce such a large anisotropy change.


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2Fourth-order uniaxial anisotropy energy is expressed as: $K_1 \sin^2 \theta + K_2 \sin^4 \theta$, whereas sixth-order cubic anisotropy energy is: $K_1 (\alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2) + K_2 (\alpha_2^2 \alpha_3^2 \alpha_1^2 \alpha_2^2 \alpha_3^2 \alpha_1^2)$ where: $\alpha_1, \alpha_2, \alpha_3$ are the cosines of the angles that the magnetization makes with the ($x, y, z$) axes, respectively (see fig. 1).
Aharoni’s approach is based on an isotropic approximation of the inhomogeneous components of the curling magnetization. Following Muller and Goldstein [15], we have developed a different set of curling equations in the presence of a fourth-order uniaxial case that is free from the isotropic approximation as well as in the presence of cubic anisotropy. In each case we get an appropriate $g(\theta)$ function as given below.

In our fourth-order uniaxial case (see footnote 2), $g(\theta)$ is given by $g(\theta) = 2(N_0 \sin^2 \theta + N_c \cos^2 \theta) - \alpha - (K_1 + 2K_2) \cos \theta - 2K_2 \sin^2 \theta (3 \sin^2 \theta - \cos^2 \theta)$.

In the sixth-order cubic anisotropy (see footnote 2) case, it is given by $g(\theta) = 2(N_0 \sin^2 \theta + N_c \cos^2 \theta) - \alpha - K_1 (1 - 5 \cos^2 \theta \sin^2 \theta) - K_2 (3 \sin^2 \theta - \cos^2 \theta)$.

The system of equations defined by $f(\theta)$ and the extraction of the $g(\theta)$ function can be used to extract the components of the switching field $h_x, h_z$ as before:

$$h_x = \sin(\theta)g(\theta) - \cos(\theta)f(\theta), h_z = \cos(\theta)g(\theta) + \sin(\theta)f(\theta).$$

We use the same parameters as Wegrowe et al. [14], $K_1 = 2.0 \times 10^5 \text{erg/cm}^3$, $K_2 = 0 \text{erg/cm}^3$ and compare them to the nickel bulk values at room temperature.

### Discussion and conclusion.
In conclusion, by means of geometrical representation and experimental measurements, we have investigated the reversal processes in ferromagnetic nanowires. Our systematic studies of the...
Two rotation modes are considered as the most important: coherent rotation and curling. Good agreement between the measured magnetic properties of Ni and Co nanowires and the theoretical calculations is obtained. Further experimental work remains to be done in order to observe this transition and finely tune its dynamics in order to exploit it efficiently in recording or in magnetic devices.

**Fig. 7:** Comparison of least-squares fitting and measurements (+) on all cobalt nanowires. Geometric signature for the Co nanowire experimental results with four sets of diameters: 15, 50, 80 and 100 nm. The fitting values are: $M_s = 87 \text{emu/cm}^3$, $K_1 = -11333 \text{erg/cm}^3$ and $K_2 = 11318 \text{erg/cm}^3$ for 15 nm, $M_s = 105 \text{emu/cm}^3$, $K_1 = -11691 \text{erg/cm}^3$ and $K_2 = 7268 \text{erg/cm}^3$ for 50 nm, $M_s = 44 \text{emu/cm}^3$, $K_1 = -9912 \text{erg/cm}^3$ and $K_2 = 22826 \text{erg/cm}^3$ for 80 nm, and $M_s = 79 \text{emu/cm}^3$, $K_1 = -90674 \text{erg/cm}^3$ and $K_2 = 28643 \text{erg/cm}^3$ for 100 nm diameter. The reduced radius is $S = 0.065, 0.262, 0.175$ and 0.396, respectively. The inset in the 100 nm case shows the detailed quality of the fit. The framing curves are obtained from the fitted $K_1$, $K_2$ and the bulk value of $M_s$.

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