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Magnetic particle embedded in a piezoelectric matrix: analysis and applications

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Abstract :

We take into consideration a nonlinear magnetostrictive particle embedded in a piezoelectric matrix in order to obtain (stress mediated) magneto electric effects with applications to memory cells developments. The micromechanical analysis is conducted through the magneto-electro-elastic Eshelby tensor in a completely anisotropic environment. The results show the equilibrium orientations of magnetization inside the particle versus the applied fields and the boundary conditions. A bi-stable behaviour (controlled by the applied electric field) is in particular demonstrated with possible applications to memory cells design.

Keywords: magnetoelectrics, magnetostrictive particle, piezoelectric materials, Eshelby theory

1 Introduction

In scientific literature there is a large interest in determining the magnetic, electric, and elastic fields in a composite with piezoelectric and magneto-ordered phases. Typically the dispersed inhomogeneities are considered as ellipsoidal inclusions ranging from thin flakes to continuous fibres. Therefore, the magneto-electro-elastic tensors analogous to the standard elastic Eshelby tensor has been introduced and largely applied for obtaining the effective properties [1]. It is found that such composites reveal interesting (stress mediated) magnetoelectric coupling which are absent in each constituent. Of course, this methodology has been applied in order to analyse the behaviour of linear composite structure, formed by linear constituents.

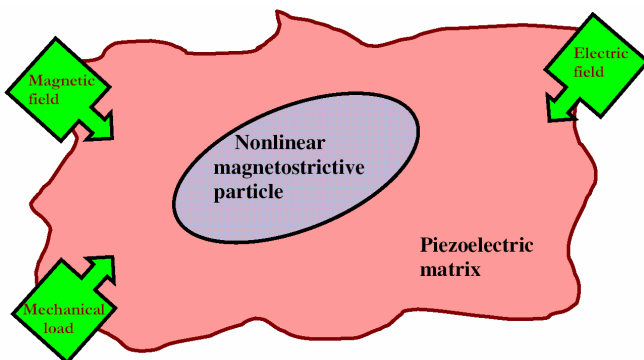


Fig. 1 Structure of the system under investigation: a nonlinear magnetostrictive particle embedded in a piezoelectric matrix. The externally applied electric, magnetic and mechanical fields are indicated and they can possibly control the magnetization orientation inside the particle.

On the other hand, numerous efforts are made in order to develop the next generation of random access memories, possibly non volatile, having low power consumption and high integration density. Recently, the different existing approaches and technologies have been compared and discussed [2]. One promising solution is based on nonlinear magnetostrictive (ferromagnetic) particles embedded in a piezoelectric matrix. Therefore, it is important to generalize the previous theories [1] in order to take into account nonlinear features of the dispersed particles [3]. In this paper, we outline a procedure able to evaluate all the physical field in the system represented in Fig. 1, where the magnetization orientation inside the particle can be obtained through ad hoc externally applied fields. In particular, a bi-stable behaviour (controlled by the applied electric field) can be obtained and it could be useful for applications to memory cells design.

2 Behaviour of the particle

To begin, we take into consideration a single particle which exhibits ferromagnetic behaviour. We suppose that the size of the particle is small enough to assure that the particle can be treated as a single ferromagnetic domain. It means that, in any case, inside the inhomogeneity an uniform magnetization appears and it is given by

$$\vec{M} = M_s \vec{\gamma} \quad (1)$$

where M_s is the constant intensity of the magnetization (saturation depending on the material under consideration) and $\vec{\gamma} = (\gamma_1, \gamma_2, \gamma_3)$ is the unknown unit vector fixing the direction of the magnetization. The principal aim of the following procedure is that of determining the orientation $\vec{\gamma}$ in terms of the externally applied fields and of the boundary conditions. It can be obtained by minimizing the energy function of the single domain particle, as follows [4,5]

$$w(\vec{\gamma}) = -\mu_0 \underbrace{M_s \vec{\gamma}}_{\vec{M}} \cdot \vec{H} + \varphi(\vec{\gamma}) - \hat{T} : \hat{\varepsilon}_\mu(\vec{\gamma}) \quad (2)$$

the first term represents the magnetic interaction energy where \vec{H} is the local magnetic field measured inside the particle: this is the Zeemann term and it describes the influence of the magnetic field on the orientation of the magnetization ($\mu_0 = 4\pi \cdot 10^{-7}$ H/m is the vacuum magnetic permeability). The second term $\varphi(\vec{\gamma})$ represents the anisotropic energy depending on the crystal class of the material. This magneto-crystalline energy tends to force the magnetization to be aligned along particular directions, called easy axes. These directions are connected to crystallographic structure and the magneto-crystalline energy is minimum when $\vec{\gamma}$ is parallel to the easy axis. The third term represents the elastic interaction energy where \hat{T} is the local stress tensor measured inside the particle and $\hat{\varepsilon}_\mu(\vec{\gamma})$ is the strain tensor induced by the magnetization (magnetostriction strain that would appear if the material was able to deform without external stress). Both \hat{T} and $\hat{\varepsilon}_\mu(\vec{\gamma})$ are uniform within the single domain particle, as discussed below.

The nonlinear minimization furnishes the direction in terms of the magnetic field and the stress tensor:

$$\min_{\vec{\gamma}: \|\vec{\gamma}\|=1} w(\vec{\gamma}; \vec{H}, \hat{T}) \Rightarrow \vec{\gamma} = \vec{\gamma}(\vec{H}, \hat{T}) \quad (3)$$

Moreover, the local magnetic field and stress tensor inside the particle depend on the environment where the particle is embedded and on the external fields applied to the structure. To conclude we can state the constitutive equations of the particle in the following way. For the magnetic point of view we have

$$\vec{B} = \mu_0 (\vec{H} + \vec{M}) = \mu_0 [\vec{H} + M_s \vec{\gamma}(\vec{H}, \hat{T})] \quad (4)$$

where \vec{B} is the magnetic induction. For the elastic point of view we have

$$\hat{T} = \hat{L}_2 \{ \hat{\varepsilon}_0 - \hat{\varepsilon}_\mu[\vec{\gamma}] \} = \hat{L}_2 \{ \hat{\varepsilon}_0 - \hat{\varepsilon}_\mu[\vec{\gamma}(\vec{H}, \hat{T})] \} \quad (5)$$

where $\hat{\varepsilon}_0$ is the local strain tensor (measured with respect to the demagnetized particle), \hat{L}_2 is the stiffness tensor of the particle (exhibiting the symmetries of the crystal under consideration).

3 Coupling with the external magnetic field

As above stated, the magnetic field \vec{H} entering the energy in Eq.(2) is the local (internal) magnetic field and, therefore, it is important to obtain its relationships with the externally applied magnetic field \vec{H}^∞ . To this aim we can utilize a recent results which is valid for an arbitrary nonlinear and anisotropic ellipsoidal particle embedded in a linear but anisotropic matrix [6]. In particular, we consider a nonlinear ellipsoidal inhomogeneity (having axes a_x , a_y and a_z) described by the (magnetic field dependent) permeability tensor $\hat{\mu}_2 = \hat{\mu}_2(\vec{H})$, embedded in a matrix with permeability tensor $\hat{\mu}_1$. In these conditions we have

$$\vec{H} = \left[\hat{I} - \hat{S}_m (\hat{I} - \hat{\mu}_1^{-1} \hat{\mu}_2) \right]^{-1} \vec{H}^\infty \quad (6)$$

where \hat{S}_m is the so-called magnetic Eshelby tensor (adimensional) given by [6]

$$\hat{S}_m = \frac{\det(\hat{a})}{2} \int_0^{+\infty} \frac{(\hat{a}^2 + s\hat{\mu}_1)^{-1} \hat{\mu}_1}{\sqrt{\det(\hat{a}^2 + s\hat{\mu}_1)}} ds \quad (7)$$

Here \hat{a} is a tensor defining the geometry of the ellipsoid: $\hat{a} = \text{diag}(a_x, a_y, a_z)$ and $\hat{a}^2 = \text{diag}(a_x^2, a_y^2, a_z^2)$. Interestingly enough, we note that the magnetic Eshelby tensor depends only on the geometry of the system and on the matrix permeability tensor. By using the definition of the nonlinear constitutive equation of the particle $\vec{B} = \hat{\mu}_2(\vec{H})\vec{H} = \mu_0(\vec{H} + \vec{M})$ we can write Eq.(6) in a different form. In fact, from Eq.(6) we obtain

$$\left[\hat{I} - \hat{S}_m (\hat{I} - \hat{\mu}_1^{-1} \hat{\mu}_2) \right] \vec{H} = \vec{H}^\infty \quad (8)$$

or, equivalently

$$\vec{H} - \hat{S}_m \vec{H} + \hat{S}_m \hat{\mu}_1^{-1} \mu_0 \vec{H} + \hat{S}_m \hat{\mu}_1^{-1} \mu_0 M_s \vec{\gamma} = \vec{H}^\infty \quad (9)$$

Finally, after straightforward calculations, we obtain

$$\vec{H} = \left[\hat{I} - \hat{S}_m (\hat{I} - \hat{\mu}_1^{-1} \mu_0) \right]^{-1} \left[\vec{H}^\infty - \hat{S}_m \hat{\mu}_1^{-1} \mu_0 M_s \vec{\gamma} \right] \quad (10)$$

It means that the local magnetic field has been explicitly written in terms of the remotely applied magnetic field and of the internal magnetization orientation

$$\vec{H} = \vec{H}(\vec{H}^\infty, \vec{\gamma}) \quad (11)$$

This is the most important achievement of this section and it will be used in the following for the development of the whole procedure. Eq.(7) can be simplified for an isotropic or crystalline cubic matrix. In such cases in fact we have $\hat{\mu}_1 = \mu_1 \hat{I}$ where μ_1 is scalar permeability and \hat{I} is the identity tensor and, therefore, we obtain the simpler version of the magnetic Eshelby tensor $\hat{S}_m = \text{diag}(L_1, L_2, L_3)$ where the

L_i 's are the so called depolarization factors $L_i = \frac{a_1 a_2 a_3}{2} \int_0^{+\infty} \frac{d\eta}{(a_i^2 + \eta) \prod_{j=1}^3 \sqrt{a_j^2 + \eta}}$ with $L_1 + L_2 + L_3 = 1$.

Moreover, for a sphere we have $a_x = a_y = a_z$ and therefore $L_1 = L_2 = L_3 = 1/3$.

4 Coupling with the external electric and elastic fields

As above introduced, the coupling with the external electric and elastic fields is mediated by the piezoelectric matrix, where the particle is embedded. We begin its analysis by defining the constitutive equations. To this aim it is useful to use the compact Voigt notation for stress and strain tensors as follows

$$\vec{T} = [T_{11}, T_{22}, T_{33}, T_{23}, T_{13}, T_{12}]^T \quad \text{and} \quad \vec{\varepsilon} = [\varepsilon_{11}, \varepsilon_{22}, \varepsilon_{33}, 2\varepsilon_{23}, 2\varepsilon_{13}, 2\varepsilon_{12}]^T \quad (12)$$

So doing, the piezoelectric behaviour can be described by the following relation (stress-charge format)

$$\begin{bmatrix} \vec{T} \\ \vec{D} \end{bmatrix} = \begin{bmatrix} \hat{L}_1 & \hat{q}_1 \\ \hat{q}_1^T & -\hat{p}_1 \end{bmatrix} \begin{bmatrix} \vec{\varepsilon} \\ -\vec{E} \end{bmatrix} \quad (13)$$

where \hat{L}_1 is the elastic stiffness tensor, \hat{q}_1 is the tensor of the piezoelectric moduli and \hat{p}_1 is the permittivity tensor of the matrix. The representation of the constitutive equation through the variables $[\vec{T}, \vec{D}]$ and $[\vec{\varepsilon}, -\vec{E}]$ is particularly indicated since always allows to a symmetric generalized stiffness tensor, composed by the objects \hat{L}_1 , \hat{q}_1 and \hat{p}_1 . Such a symmetry can be proved by thermodynamic-energetic arguments [7]. Alternatively, a different notation can be adopted by defining the following generalized variables

$$\hat{\Sigma} = \left[\hat{T} \middle| \vec{D} \right] \quad \text{and} \quad \hat{Z} = \begin{bmatrix} \hat{\varepsilon} \\ -\vec{E} \end{bmatrix} \quad (14)$$

where $\hat{\Sigma}$ is represented by a matrix 3x4 and \hat{Z} is represented by a matrix 4x3. Therefore, the matrix behaviour can be summarized through the relation

$$\Sigma_{ij} = (\Lambda_1)_{iJMn} Z_{Mn} \quad (15)$$

where $i, n=1..3$ and $J, M=1..4$. The tensor $\hat{\Lambda}_1$ represents the overall piezoelectric behaviour of the matrix material and contains all the components \hat{L}_1, \hat{q}_1 and \hat{p}_1 . Now, it is important to observe that the particle is embedded into the matrix when it has a specific magnetization state identified by an initial direction $\vec{\gamma}_0$. We want to measure the strain with respect to such a configuration and, therefore, we define the local strain as $\hat{\epsilon} = \hat{\epsilon}_0 - \hat{\epsilon}_\mu(\vec{\gamma}_0)$ where $\hat{\epsilon}_0$ is defined in Section 2.

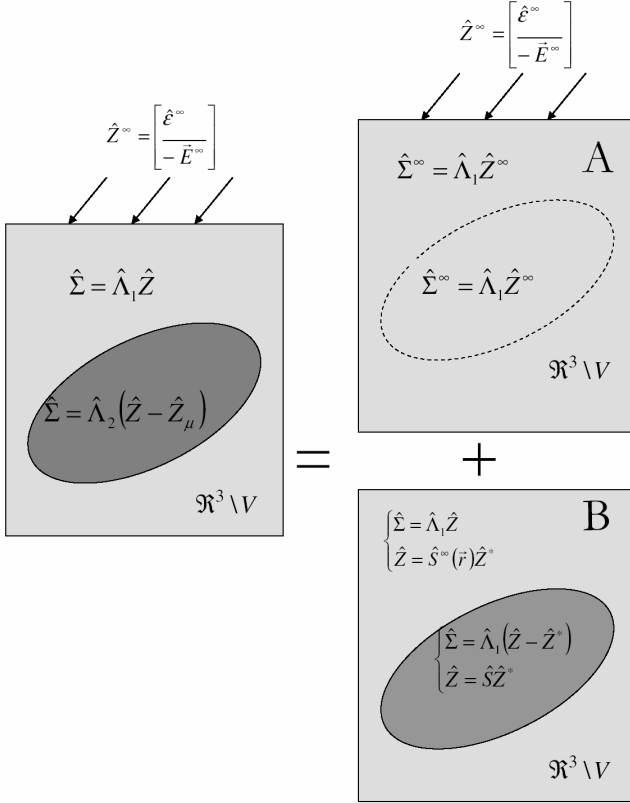


Fig. 2 Scheme describing the Eshelby equivalence principle applied to the case of a piezoelectric matrix. The problem A corresponds to a uniform medium $\hat{\Lambda}_1$ with remotely applied fields \vec{E}^∞ and $\hat{\epsilon}^\infty$. In such a case we observe that the fields remain uniform in the entire space. The problem B represents an inclusion problem with eigenfield \hat{Z}^* and without external actions. In this case the generalized strain and stress are given by $\hat{Z} = \hat{S} \hat{Z}^*$ and $\hat{\Sigma} = \hat{\Lambda}_1 (\hat{S} - \hat{I}) \hat{Z}^*$, as predicted by the Eshelby theory. The superimposition of the two sub-problems allows us to solve the original problem of the particle described by $\hat{\Sigma} = \hat{\Lambda}_2 (\hat{Z} - \hat{Z}_\mu)$ embedded in the piezoelectric matrix.

The same representation of Eq.(13) can be applied to the elastic-dielectric response of the particle. In this case we obtain

$$\begin{bmatrix} \vec{T} \\ \dots \\ \vec{D} \end{bmatrix} = \begin{bmatrix} \hat{L}_2 & \hat{0} \\ \hat{0} & -\hat{p}_2 \end{bmatrix} \left(\begin{bmatrix} \vec{\epsilon} \\ \dots \\ -\vec{E} \end{bmatrix} - \begin{bmatrix} \vec{\epsilon}_\mu(\vec{\gamma}) - \vec{\epsilon}_\mu(\vec{\gamma}_0) \\ \dots \\ \vec{0} \end{bmatrix} \right) \quad (16)$$

where \hat{L}_2 and \hat{p}_2 are the elastic stiffness and the permittivity tensor of the particle, respectively (for a metallic behaviour of the particle we can set $\hat{p}_2 = p \hat{I}$ with p approaching infinity). The vector $\vec{\epsilon}_\mu(\vec{\gamma})$ represents the Voigt form of the magnetostrictive strain $\hat{\epsilon}_\mu(\vec{\gamma})$. As before, such a constitutive equation can be also written through the variables defined in Eq.(14), by obtaining

$$\Sigma_{ij} = (\Lambda_2)_{iJMn} (Z_{Mn} - Z_{\mu Mn}) \quad (17)$$

where $i, n=1..3$ and $J, M=1..4$ and the tensor $\hat{\Lambda}_2$ represents the behaviour of the particle material, containing the components \hat{L}_2 and \hat{p}_2 . Here $Z_{\mu Mn}$ is the vertical juxtaposition of $\vec{\epsilon} = \vec{\epsilon}_\mu(\vec{\gamma}) - \vec{\epsilon}_\mu(\vec{\gamma}_0)$ and $-\vec{E} = \vec{0}$, as defined in Eq.(14). The analysis of this configuration can be conducted by applying the Eshelby equivalence principle. We suppose that the whole structure is subjected to an external uniform electric field \vec{E}^∞ and a remote uniform strain $\hat{\epsilon}^\infty$. We are searching for the perturbation to these uniform fields induced by the presence of the inhomogeneity. The equivalence principle, which we are going to illustrate, has been summarized in Fig. 2. The actual presence of an inhomogeneity can be described by the superimposition of

the effects generated by two different situations A and B. The first situation is very simple because it considers the effects of the remote fields in an homogeneous matrix without the inhomogeneity. The situation B corresponds to an inclusion scheme (eigenfield uniformly distributed within the particle) where the eigenfield \hat{Z}^* is still unknown and it can be determined by imposing the equivalence between the original problem and the superimposition A+B. The total fields inside the particle can be obtained summing up the two contributions A and B as follows

$$\begin{aligned}\hat{Z} &= \hat{Z}^\infty + \hat{S}\hat{Z}^* \\ \hat{\Sigma} &= \hat{\Lambda}_1\hat{Z}^\infty + \hat{\Lambda}_1(\hat{S} - \hat{I})\hat{Z}^*\end{aligned}\quad (18)$$

where the eigenfield \hat{Z}^* must be obtained imposing the correct behavior of the particle, which means its constitutive equation

$$\hat{\Sigma} = \hat{\Lambda}_2(\hat{Z} - \hat{Z}_\mu) \quad (19)$$

By substituting Eq.(18) in Eq.(19) we obtain the tensor equation for \hat{Z}^*

$$\hat{\Lambda}_1\hat{Z}^\infty + \hat{\Lambda}_1(\hat{S} - \hat{I})\hat{Z}^* = \hat{\Lambda}_2(\hat{Z}^\infty + \hat{S}\hat{Z}^* - \hat{Z}_\mu) \quad (20)$$

By solving Eq.(20) we straightforwardly obtain the equivalent eigenfield

$$\hat{Z}^* = \left[(\hat{I} - \hat{\Lambda}_1^{-1}\hat{\Lambda}_2) - \hat{S} \right]^{-1} \left[\hat{Z}^\infty - (\hat{I} - \hat{\Lambda}_2^{-1}\hat{\Lambda}_1) \hat{Z}_\mu \right] \quad (21)$$

and, therefore, Eq.(18) (coupled with Eq.(21)) give us the complete solutions of the problem. As result, the local elastic stress depends on the external elastic and electric fields and on the magnetization direction through a function which is now available. It can be summarized as

$$\hat{T} = \hat{T}(\hat{\epsilon}^\infty, \vec{E}^\infty, \vec{\gamma}) \quad (22)$$

This is the most important result of the present section and it will be used to define the complete procedure discussed in the following. The Eshelby tensor, which appears in the previous calculations, is defined in such a way to describe the response of an inclusion problem (with a given eigenfield), as stated in the above defined sub-problem B. A very large amount of literature has been devoted to its evaluation and its properties [1,8,9].

5 Complete system of equations

The set of equations describing the magnetoelastic particle embedded into the piezoelectric matrix is given by the energy minimisation for the particle $\vec{\gamma} = \vec{\gamma}(\vec{H}, \hat{T})$ (see Eq.(3)), by the coupling with the external magnetic field $\vec{H} = \vec{H}(\vec{H}^\infty, \vec{\gamma})$ (see Eq.(11)) and by the coupling with external elastic and electric fields $\hat{T} = \hat{T}(\hat{\epsilon}^\infty, \vec{E}^\infty, \vec{\gamma})$ (see Eq.(22)). The problem is well posed: the three unknowns \hat{T} , \vec{H} and $\vec{\gamma}$ can be found when the external fields $\hat{\epsilon}^\infty$, \vec{E}^∞ and \vec{H}^∞ are given.

The problem can be simplified by rewriting the first equation in terms of the derivatives of w and by applying the method of the Lagrangian multipliers in order to take into account the constraint $\|\vec{\gamma}\| = 1$

$$\frac{\partial}{\partial \gamma_i} \left[w(\vec{\gamma}, \vec{H}, \hat{T}) - \lambda(\vec{\gamma} \cdot \vec{\gamma} - 1) \right] = 0 \Rightarrow -\mu_0 M_s H_i + \frac{\partial \varphi(\vec{\gamma})}{\partial \gamma_i} - \hat{T} : \frac{\partial \hat{\epsilon}_\mu(\vec{\gamma})}{\partial \gamma_i} = 2\lambda \gamma_i \quad (23)$$

We can also exploit the linear dependences in the other two relations

$$\begin{aligned}\vec{H} &= \vec{H}(\vec{H}^\infty, \vec{\gamma}) = \hat{A}\vec{H}^\infty + \hat{B}\vec{\gamma} \\ \hat{T} &= \hat{T}(\hat{\epsilon}^\infty, \vec{E}^\infty, \vec{\gamma}) = \hat{C}\hat{\epsilon}^\infty + \hat{D}\vec{E}^\infty + \hat{F}[\hat{\epsilon}_\mu(\vec{\gamma}) - \hat{\epsilon}_\mu(\vec{\gamma}_0)]\end{aligned}\quad (24)$$

where the tensors \hat{A} , \hat{B} , \hat{C} , \hat{D} and \hat{F} can be obtained through the procedures outlined in the previous sections. The problem can be converted to a single minimization problem as follow. We substitute Eq.(24) in Eq.(23) and, since the tensors \hat{B} and \hat{F} are symmetric, we can define a new energy function \tilde{w} as follows

$$\begin{aligned}
\tilde{w} = & \underbrace{-\mu_0 M_s \vec{\gamma} \cdot \hat{A} \vec{H}^\infty - \frac{1}{2} \mu_0 M_s \vec{\gamma} \cdot \hat{B} \vec{\gamma}}_{\text{magnetic}} + \underbrace{\varphi(\vec{\gamma})}_{\text{anisotropic}} \\
& \underbrace{-\hat{C} \hat{\varepsilon}^\infty : \hat{\varepsilon}_\mu(\vec{\gamma}) - \hat{D} \vec{E}^\infty : \hat{\varepsilon}_\mu(\vec{\gamma}) - \frac{1}{2} \hat{F} \hat{\varepsilon}_\mu(\vec{\gamma}) : \hat{\varepsilon}_\mu(\vec{\gamma}) + \hat{F} \hat{\varepsilon}_\mu(\vec{\gamma}_0) : \hat{\varepsilon}_\mu(\vec{\gamma})}_{\text{elastic}}
\end{aligned} \quad (25)$$

So, the initial minimization problem stated in Eq.(5) has been converted to the following one

$$\min_{\vec{\gamma}: \|\vec{\gamma}\|=1} \tilde{w}(\vec{\gamma}; \vec{H}^\infty, \hat{\varepsilon}^\infty, \vec{E}^\infty) \Rightarrow \vec{\gamma} = \vec{\gamma}(\vec{H}^\infty, \hat{\varepsilon}^\infty, \vec{E}^\infty) \quad (26)$$

which is much more convenient since it leads to the final magnetization orientation directly in terms of the external fields applied to the structure. In other words, the minimum of the energy function \tilde{w} represents the real orientation of the magnetization when all the external field are fixed. An example of implementation of Eq. (25) can be obtained for a two-dimensional system of possible interest for applications in memory cells design. It is described by the function $\tilde{w} = \tilde{w}(\varphi)$ where φ is the orientation angle of the magnetization on the plane. It is composed by an elliptic particle exhibiting uniaxial anisotropy [10,11] with semi-axes a_x (easy-axis) and $a_y < a_x$ (hard-axis) aligned to the reference frame. We apply an external magnetic field fixed (in the direction of the y-axis, $\varphi = \pi/2$) and an external electric field with intensity varying within a given range (in the fixed direction $\varphi = \pi/4$) [12]. The anisotropic and magnetic terms are independent of the electric field intensity, while the elastic and total terms are strongly depending on it. As result, it is possible to prove that the electric field controls the orientation of the magnetization between two different states, generating a bistable behaviour very useful for applications in memory devices [12,13]. The details of this result will be published elsewhere in the near future.

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