Current-induced switching and magnetoresistance of noncollinear bulk magnetic structures

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(Received 31 May 2010; revised manuscript received 29 July 2010; published 13 September 2010)

In our work, we theoretically demonstrated the possibility to control magnetic configurations and magnetoresistance of a wide class of magnetically frustrated materials with applied voltages. This phenomenon may be viewed as the bulk material counterpart of spin-transfer torque and may represent a critical interest both from applications and fundamental points of view. It is shown that the magnetic configuration of bulk magnets strongly depends on position of the Fermi level and applied voltage. We propose this phenomenon for future experimental studies suggesting materials with a strong variation in density of states near the Fermi level for different magnetic configurations.

DOI: 10.1103/PhysRevB.82.094420 PACS number(s): 75.47.Pq, 72.15.Gd, 75.10.Jm, 75.10.Lp

I. INTRODUCTION

The discovery of giant magnetoresistance (GMR) (Refs. 1 and 2) in magnetic multilayered structures has generated a new field of spin-based electronics[3,4] or spintronics, which combines two traditional fields of physics: magnetism and electronics. A spin-valve concept[5] used in GMR structures allows controlling the magnetic configuration of its ferromagnetic layers by (i) application of relatively small magnetic fields or (ii) passing spin-polarized currents using spin-transfer torque (STT).[6,7] These factors made them ideal systems for spintronic applications such as magnetic random access memories and magnetic field sensors used in read heads.

The advent of GMR has considerably increased an interest in related phenomenon in bulk materials, colossal magnetoresistance (CMR),[8,9] which is several orders higher than GMR and unlike the latter, can be viewed as an "intrinsic" property of material itself. To date, the CMR is typically observed in certain manganite compounds with the bulk magnetic configuration controlled by applying the magnetic field [similar to method (i) mentioned above for spin valves] but requires characteristic magnetic fields of several tesla.[9] Such high fields make them inappropriate for use in spintronic applications where appropriate scale should be about oersteds. However, one may expect the possibility of controlling the intrinsic magnetic configuration of the bulk materials (and thus of CMR) by passing spin-polarized currents through them similar to STT mechanism (ii) mentioned above for spin valves. Since the STT in the latter originates from noncollinearity of their adjacent magnetizations, the same requirement should hold for magnetic moments in the bulk materials.

Here we promote magnetically frustrated bulk materials as a new paradigm for spintronic applications with high magnetoresistance which can be controlled with relatively small applied voltages and does not require injection of spin-polarized currents. This phenomenon may be viewed as the "bulk" counterpart of STT in layered spintronic structures (spin valves) and may represent a crucial interest both from applications and fundamental points of view. Below we demonstrate that the magnetic configuration of the bulk frustrated material is changed under applied voltage leading to the strong variation in its conductance.

The key mechanism at stake is a somewhat microscopic equivalent of spin torque, allowing for local spin flips within the microscopic spin texture. Those local moves require noncollinearity of the spin configurations, which would otherwise be insensitive to the current. In order to demonstrate the possibility of switching between different magnetically ordered configurations, we design a toy model which, for a given set of parameters, always has a unique noncollinear ground state. Noncollinearity is achieved by introducing multiaxial anisotropies for the localized moments. When voltage is applied, this model will evolve to different well-defined noncollinear ground states, similar to a spin model which evolves from one magnetic configuration to another one through application of a magnetic field. The details of the switching dynamics are beyond the scope of this work as this aspect will be specific to each material. In addition, similarly to the case of GMR,[1,2] we do not consider the bistability issue because competition between different ground states occurs at the microscopic level and once the nonequilibrium current is shut down, the model goes back to its initial magnetic configuration.

II. MODEL

We consider a square lattice of classical localized moments $\mathbf{S}_i$ with strong local on-site uniaxial anisotropy (D_0) along square diagonals (represented by unit vector $\mathbf{n}_i$) and coupled through intersite exchange I_{ij}. In addition, moments $\mathbf{S}_i$ are coupled with conduction electrons through the local exchange (J_0) described here quantum mechanically using tight-binding model. Of note, since exchange interaction involves classical localized moments our description remains to be a single-electron problem. Unlike collinear systems, noncollinearity provided by magnetic frustration is a key ingredient for switching phenomenon proposed here since its origin is due to STT mechanism acting locally by conduction electrons on localized moments $\mathbf{S}_i$. The Hamiltonian of the system has the form

$$\hat{H} = -\sum_{i,j} I_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - D_0 \sum_i (\boldsymbol{n}_i \cdot \boldsymbol{S}_i)^2 + t \sum_{i,j,\sigma} (\hat{c}_i^{\dagger\sigma} \hat{c}_j^{\sigma} + \text{H.c.}) - J_0 \sum_i \hat{c}_i^{\dagger\alpha} (\boldsymbol{\sigma}_{\alpha\beta} \cdot \boldsymbol{S}_i) \hat{c}_i^{\beta}, \quad (1)$$

where I_{ij} and J_0 are the intersite and the local exchange constants, respectively, D_0 is the uniaxial anisotropy constant, t is the hopping integral between two neighboring sites, $\hat{c}_i^{\dagger\sigma}$ and $\hat{c}_i^{\sigma}$ are the creation and annihilation operators of the conduction electron with the spin σ on site i, and $\boldsymbol{\sigma}_{\alpha\beta}$ is the vector of Pauli matrices. For the chosen model, nearest-neighbor interactions I_1 are irrelevant (because nearest-neighbor spins are always orthogonal) so that only the second-nearest neighbor one I_2 is taken into account. We emphasize that dealing with longer ranges interaction does not affect further results reported here since only energy differences between magnetic configurations matter.

Even though we use a toy model to demonstrate a concept, realistic realizations would involve more complicated structures. Several multiaxial anisotropic rare-earth-transition-metal intermetallics systems are relevant candidates: in these materials frustration arises due to the competition of crystal-field anisotropy, exchange and quadrupolar interactions (for a review see Ref. 10). These intermetallics systems often show a noncollinear magnetic structure: this is the case, for example, of $TbGa_2$,[11] $HoGe_3$,[12] uranium compounds,[13] and transition-metal-based compounds such as MnSi,[14–16] MnIrSi,[17,18] FeMn,[19,20] $FeGe_2$,[21] and $PdCrO_2$.[22] Naturally, pyrochlores in which frustration is due to the crystal structure are also relevant candidates, as it is well known that in such exotic systems, noncollinear low temperature magnetic phases are often stabilized,[23] among those, few metallic compounds have been identified such as $Pr_2Ir_2O_7$ (Refs. 24 and 25) and the family of the pyrochlore Molybdates, $R_2Mo_2O_7$.[26] Clearly, each of these systems should be described by appropriate microscopic Hamiltonians in order to fully take into account all magnetic properties, including the effect of anisotropies, noncollinearity of the ground states, ordering temperatures, etc.

Coming back to the model Hamiltonian (1), in the limit of low temperature and $D_0 \to \infty$, the localized moments $\mathbf{S}_i$ are strictly collinear with $\mathbf{n}_i (\mathbf{S}_i \| \mathbf{n}_i)$ and the total energy of the system within a constant shift becomes

$$E_{tot} = -I_2 \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + \text{Tr}[\hat{H}\hat{\rho}] = -I_2 \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j - \frac{i}{2\pi} \sum_j \int E G_{jj}^{<}(E) dE, \quad (2)$$

where $G^{<}$ and $\hat{\rho}$ represent the "lesser" nonequilibrium Green's function and density matrix, respectively,[27] and the Green's function indices include spin index as well.

We adopt a conventional transport approach where the system is considered to consist of three regions: semi-infinite left (L) and right (R) regions connected to the middle scattering region (M) which is assumed to be very long. Despite of all three regions being identical, such a subdivision is chosen in order to consider all nonequilibrium processes occurring in the scattering region M while the left and right regions are in thermodynamical equilibrium described with Fermi-Dirac distribution functions $f_{L(R)} = f(E - \mu_{L(R)})$, where $\mu_{L(R)}$ is the chemical potential in the left (right) region. Omitting spin indexes, the lesser Green's function can be written as

$$G_{ij}^{<} = i(f_L \psi_i^L \psi_j^{L*} + f_R \psi_i^R \psi_j^{R*}), \quad (3)$$

where $\psi_j^{L(R)}$ is the single-electron wave function on site $j \in M$ incident from the left (right). It is straightforward from the definition of retarded and advanced Green's functions that

$$G_{jj}^{a} - G_{jj}^{r} = i(\psi_j^L \psi_j^{L*} + \psi_j^R \psi_j^{R*}). \quad (4)$$

Since the middle region is assumed to be very long, the finite potential drop $eV = \mu_L - \mu_R$ results into infinitesimal change from one unit cell to another within the middle region M. In this case the reflections from the $L|M$ and $R|M$ boundaries are negligible and one can write for the left (right) wave functions $\psi_j^{L(R)} = c_{L(R)} \exp[+(-) k_x a j]$, where k_x is the wave vector along x axis and a is the lattice constant. One can show that the expression for the charge current which is proportional to $(f_L - f_R)$ (Ref. 28) is satisfied when $|\psi_j^L|^2 = |\psi_j^R|^2$. Indeed, the charge current can be expressed using the lesser Green's function as[29]

$$I \sim \int [(G_{j+1,j}^{<} - G_{j,j+1}^{<})] dE.$$

Using Eq. (3) and aforementioned wave functions, it yields the following form:

$$I \sim \int \sin k_x a [f_L |c_L|^2 - f_R |c_R|^2] dE$$

which is proportional to $(f_L - f_R)$ only when $|c_L|^2 = |c_R|^2$ (and consequently $|\psi_j^L|^2 = |\psi_j^R|^2$).

Then, it straightforwardly follows from Eqs. (3) and (4) that

$$G_{jj}^{<}(E) = i(f_L + f_R) \Im[G_{jj}^{r}(E)], \quad (5)$$

where $\Im$ notation for imaginary part is used. The total energy given by expression (2) of the system per a unit cell yields

$$E_{tot} = -I_2 \sum_{i,j} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - \frac{1}{2\pi} \int (f_L + f_R) \sum_j \Im[G_{jj}^{r}(E)] E dE, \quad (6)$$

where $G_{jj}^{r}(E)$ is calculated as jth diagonal element of the matrix $[E - \hat{H} + i\delta]^{-1}$. Thus, the magnetic configuration corresponding to the minimum of the total energy is resulting from the interplay of intersite exchange interaction between localized moments on the next-nearest-neighbor sites [the first term in Eq. (6)] and the exchange interaction between the localized moment and the conduction electron described by the second term in Eq. (6).

For simplicity, we consider a four-site unit cell with three possible magnetic configurations of the system $(\mathbf{S}_i \| \mathbf{n}_i)$: "4in," "3in1out," and "2in2out" (see Fig. 1). It is sufficient considering these configurations only since other possible

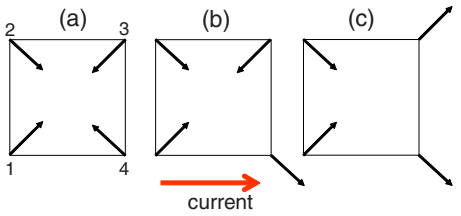

FIG. 1. (Color online) (a) "4in," (b) "3in1out," and (c) "2in2out" configurations for square lattice.

configurations, such as "4out," "3out1in," and "2out2in" are equivalent to the chosen ones depending on a choice of the unit cell. We choose for parameters $J_0=2$ eV, $t=1$ eV, and $I_2=0.1t$.[30]

III. RESULTS AND DISCUSSION

In Fig. 2, we show the dependence of the total energy for the three aforementioned selected configurations as a function of applied voltage. First, one can note that E_{tot} strongly depends on the Fermi-level position which is used to represent different materials. Indeed, when the system is in equilibrium, its "2in2out" ("4in") state is energetically more favorable for $E_F=-2$ eV (-2.5 eV) as shown in Figs. 2(a) and 2(b), respectively. Furthermore, the strong variation in the total energy as a function of applied voltage causes the system to switch from an equilibrium state to another one at certain critical voltage (see Fig. 2) with "3in1out" intermediate state stabilized at intermediate voltages. Moreover, further increase in the applied voltage may again reverse the state of the system [Fig. 2(a)]. This demonstrates that the magnetic configuration of the system can be controlled by the applied voltage.

The mechanism of these dependences may be understood from the corresponding total density of states (DOS) (Ref. 31) for considered three configurations represented in Fig. 3 where for convenience purposes only negative energy range

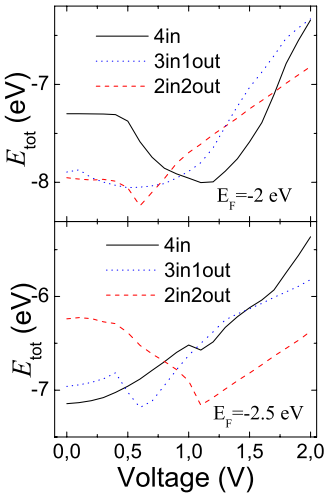

FIG. 2. (Color online) (a) and (b) Total energy as a function of applied voltage for $E_F=-2$ eV and $E_F=-2.5$ eV, respectively, shown by arrows in Fig. 3.

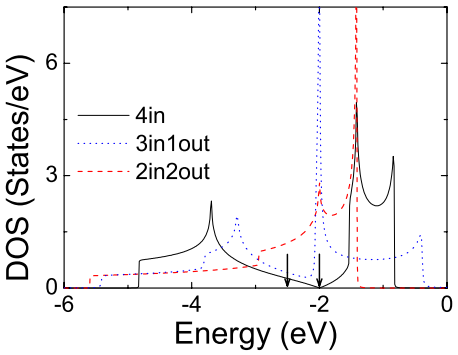

FIG. 3. (Color online) Total density of states for "4in" (solid), "3in1out" (dotted), and "2in2out" (dashed) configurations represented in Fig. 1.

is shown since $\mathrm{DOS}(E)=\mathrm{DOS}(-E)$. The DOS for all magnetic states have sharp peaks and band gaps causing strong dependence of the preferable configuration on the Fermi level E_F both in and out of equilibrium. Indeed, as on can see from the second term in Eq. (6), the total energy is defined by the sum of two products of the DOS with E integrated till μ_L and μ_R. In the absence of applied voltage, E_{tot} is calculated with $\mu_L=\mu_R=E_F$, i.e., defined by the position of the Fermi level indicated by black arrows in Fig. 3 corresponding to two values -2 eV and -2.5 eV used in Figs. 2(a) and 2(b), respectively.

One might expect that the conductances of two aforementioned magnetic configurations may strongly differ due to significant differences in their DOS, thereby leading to high magnetoresistance values defined as $(\sigma_{xx}^{2in2out}-\sigma_{xx}^{4in})/\sigma_{xx}^{4in}$, where σ_{xx}^{4in} and $\sigma_{xx}^{2in2out}$ are the conductances for "4in" and "2in2out" outermost magnetic configurations, respectively. We calculate the linear response conductance using the Kubo formula[32]

$$\sigma_{xx}=\frac{\pi e^2\hbar}{L}\mathrm{Tr}[\hat{v}_x\delta(E-\hat{H})\hat{v}_x\delta(E-\hat{H})],$$

where $\hat{v}_x$ is the electron velocity operator along the applied voltage direction, $L\to\infty$ is the length of the system, and

$$\delta(E-\hat{H})=-\frac{1}{\pi}\Im[\hat{G}^r(E)]=\frac{i}{2\pi}[\hat{G}^r(E)-\hat{G}^a(E)].$$

The calculated conductance of the system for three considered magnetic configurations are represented in Fig. 4. As one can see, conductances strongly depend on position of the Fermi energy E_F and strongly correlate with DOS picture (see Fig. 3). For the two values of the Fermi energy used above, the MR ratio is found to be about 336% for the case when $E_F=-2.5$ eV and becomes infinite for $E_F=-2$ eV.

One can note that similar behavior is expected to occur in systems with collinear antiferromagnetic (AF) structure: depending on the band structure and the band filling, the ferromagnetic state can be close in energy with the AF one, and an applied voltage could stabilize the ferromagnetic state. However in such a case, there is no spin torque since the structure is colinear, and no mechanism for spin reversal (except through spin waves). On the contrary, the mechanism

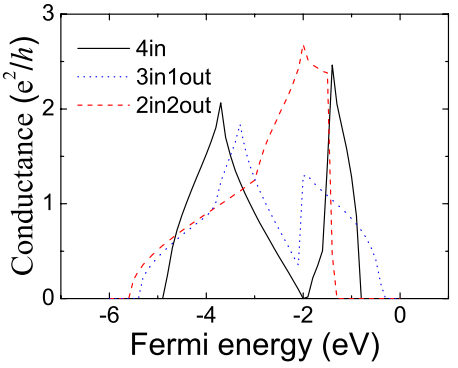

FIG. 4. (Color online) Conductance as a function of the Fermi level for configurations represented in Fig. 1.

proposed here neither require any defect, spin wave nor a fine tuning of any kind. The noncollinear nature of the magnetic configurations considered provides a robust mechanism for microscopic magnetic switching. Furthermore, this mechanism does not require spin-polarized current injection.

In conclusion, we have shown that magnetic configurations of noncollinear magnets can be controlled by applied voltage. The proposed phenomenon is the bulk material analog of spin-transfer torque used in layered spin-valve structures. In addition, magnetoresistance of these materials may reach extremely high values which open the possibility for their use in spintronics. We believe that our work will stimulate experimental studies aiming to find the proposed mechanism in appropriate materials. In the long term, in combination with inverse approach of designing the crystal structure from predefined electronic-structure properties,[33] this will open additional opportunities to expand this phenomenon to alternative classes of materials.

ACKNOWLEDGMENTS

We thank B. Dieny, A. Schuhl, and W. H. Butler for fruitful discussions. This work was supported by Chair of Excellence Program of the Nanosciences Foundation in Grenoble, France.

[1] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).
[2] G. Binasch, P. Grünberg, F. Saurenbach, and W. Zinn, Phys. Rev. B **39**, 4828 (1989).
[3] A. Fert, A. Barthelemy, J. Ben Youssef, J. P. Contour, V. Cros, J. M. De Teresa, A. Hamzic, J. M. George, G. Faini, J. Grollier, H. Jaffres, H. Le Gall, F. Montaigne, F. Pailloux, and F. Petroff, Mater. Sci. Eng., B **84**, 1 (2001).
[4] S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnar, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).
[5] B. Dieny, V. S. Speriosu, S. S. P. Parkin, B. A. Gurney, D. R. Wilhoit, and D. Mauri, Phys. Rev. B **43**, 1297 (1991).
[6] J. Slonczewski, J. Magn. Magn. Mater. **159**, L1 (1996).
[7] L. Berger, Phys. Rev. B **54**, 9353 (1996).
[8] R. von Helmolt, J. Wecker, B. Holzapfel, L. Schultz, and K. Samwer, Phys. Rev. Lett. **71**, 2331 (1993).
[9] S. Jin, T. H. Tiefel, M. Mccormack, R. A. Fastnacht, R. Ramesh, and L. H. Chen, Science **264**, 413 (1994).
[10] D. Gignoux and D. Schmitt, J. Magn. Magn. Mater. **100**, 99 (1991); J. Alloys Compd. and **326**, 143 (2001).
[11] I. Auneau, G. L. F. Fraga, D. Gignoux, D. Schmitt, and F. Y. Zhang, Physica B **212**, 351 (1995).
[12] P. Schobinger-Papamantellos, J. Rodriguez-Carvajal, L. D. Tung, C. Ritter, and K. H. J. Buschow, J. Phys.: Condens. Matter **20**, 195202 (2008).
[13] P. Burlet, J. Rossatmignod, R. Troc, and Z. Henkie, Solid State Commun. **39**, 745 (1981).
[14] G. Shirane, R. Cowley, C. Majkrzak, J. B. Sokoloff, B. Pagonis, C. H. Perry, and Y. Ishikawa, Phys. Rev. B **28**, 6251 (1983).
[15] K. Koyama, T. Goto, T. Kanomata, and R. Note, Phys. Rev. B **62**, 986 (2000).
[16] M. Lee, W. Kang, Y. Onose, Y. Tokura, and N. P. Ong, Phys. Rev. Lett. **102**, 186601 (2009).
[17] T. Eriksson, R. Lizárraga, S. Felton, L. Bergqvist, Y. Andersson, P. Nordblad, and O. Eriksson, Phys. Rev. B **69**, 054422 (2004).
[18] T. Eriksson, L. Bergqvist, T. Burkert, S. Felton, R. Tellgren, P. Nordblad, O. Eriksson, and Y. Andersson, Phys. Rev. B **71**, 174420 (2005).
[19] A. Sakuma, J. Phys. Soc. Jpn. **69**, 3072 (2000).
[20] K. Nakamura, T. Ito, A. J. Freeman, L. Zhong, and J. Fernandez-de-Castro, Phys. Rev. B **67**, 014405 (2003).
[21] L. M. Corliss, J. M. Hastings, W. Kunnmann, R. Thomas, J. Zhuang, R. Butera, and D. Mukamel, Phys. Rev. B **31**, 4337 (1985).
[22] H. Takatsu, H. Yoshizawa, S. Yonezawa, and Y. Maeno, Phys. Rev. B **79**, 104424 (2009).
[23] J. Gardner, M. J. P. Gingras, and J. E. Greedan, Rev. Mod. Phys. **82**, 53 (2010).
[24] S. Nakatsuji, Y. Machida, Y. Maeno, T. Tayama, T. Sakakibara, J. van Duijn, L. Balicas, J. N. Millican, R. T. Macaluso, and J. Y. Chan, Phys. Rev. Lett. **96**, 087204 (2006).
[25] Y. Machida, S. Nakatsuji, Y. Maeno, T. Tayama, T. Sakakibara, and S. Onoda, Phys. Rev. Lett. **98**, 057203 (2007).
[26] I. Kézsmárki, N. Hanasaki, D. Hashimoto, S. Iguchi, Y. Taguchi, S. Miyasaka, and Y. Tokura, Phys. Rev. Lett. **93**, 266401 (2004).
[27] E. M. Lifshitz and L. P. Pitaevskii, *Physical Kinetics*, Course of Theoretical Physics (Pergamon, Oxford, 1981), Vol. 10.
[28] Y. Meir and N. S. Wingreen, Phys. Rev. Lett. **68**, 2512 (1992).
[29] C. Caroli, R. Combescot, P. Nozieres, and D. Saint-James, J. Phys. C **4**, 916 (1971).
[30] I. V. Solovyev, Phys. Rev. B **67**, 174406 (2003).
[31] The density of states is equal to $-1/\pi\Sigma_j \Im G^R_{jj}(E)$, where summation includes all atoms in the unit cell and spins.
[32] R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957).
[33] A. Franceschetti and A. Zunger, Nature (London) **402**, 60 (1999).