APL Materials

Magnetoelectric materials, phenomena, and devices

Cite as: APL Mater. **9**, 050401 (2021); https://doi.org/10.1063/5.0053631
Submitted: 09 April 2021 • Accepted: 11 April 2021 • Published Online: 03 May 2021

Liza Herrera Diez, Robert Kruk, Karin Leistner, et al.

Magnetoelectric materials, phenomena, and devices

Cite as: APL Mater. 9, 050401 (2021); doi: 10.1063/5.0053631
Submitted: 9 April 2021 •
Published Online: 3 May 2021

Liza Herrera Diez,[1,a)] Robert Kruk,[2,b)] Karin Leistner,[3,4,c)] and Jordi Sort[5,6,d)]

AFFILIATIONS

[1] Centre de Nanosciences et de Nanotechnologies, CNRS, Universite Paris-Saclay, 91120 Palaiseau, France
[2] Institute of Nanotechnology, Karlsruhe Institute of Technology (KIT), 76344 Eggenstein-Leopoldshafen, Germany
[3] Leibniz IFW Dresden, Helmholtzstrasse 20, Dresden 01069, Germany
[4] Faculty of Natural Sciences, Institute of Chemistry, Chemnitz University of Technology, Straße der Nationen 62, 09111 Chemnitz, Germany
[5] Departament de Física, Universitat Autònoma de Barcelona, E-08193 Cerdanyola del Vallès, Spain
[6] Institució Catalana de Recerca i Estudis Avançats (ICREA), Pg. Lluís Companys 23, E-08010 Barcelona, Spain

Note: This paper is part of the Special Topic on Magnetoelectric Materials, Phenomena, and Devices.
[a)] **Author to whom correspondence should be addressed:** liza.herrera-diez@c2n.upsaclay.fr
[b)] **Electronic mail:** robert.kruk@kit.edu
[c)] **Electronic mail:** K.Leistner@ifw-dresden.de
[d)] **Electronic mail:** Jordi.Sort@uab.cat

https://doi.org/10.1063/5.0053631

Progress in information and communication technologies largely relies on an optimized utilization of electric power. Nanomagnetism and spintronics have largely contributed to the digital revolution by drastically increasing hard disk capacity and data processing speeds. Innovative energy-saving solutions are also sought in magnetic actuation technologies, which are at the heart of micro/nano-electro-mechanical systems (MEMS/NEMS) and many other engineering applications. However, in both cases, either to write magnetic bits of data or to operate small-sized magnetic systems, miniaturized electromagnets are required. The utilization of electric currents in such electromagnets (or spin-torque effects in spintronics) involves a significant power dissipation due to the Joule heating effect. This is aggravated at the micro-/nanoscales, where the small device dimensions hinder heat conduction and thus effective cooling. As a result, roughly half of the electric power reaching large data servers is wasted in the form of heat dissipation or in sophisticated cooling systems.[1] In view of enhancing energy efficiency, the last two decades have seen a renaissance of the interest for controlling magnetism with electrical voltage. The so-called "converse magnetoelectric effect" has emerged as a fundamentally different magnetic control and actuation approach, prompting the development of ultralow power consumption devices operated at the nanoscale.

Electricity and magnetism have always had an intimate link. There are various ways to manipulate magnetism with voltage:[2–4] (i) using single-phase multiferroics, where various types of ferroic orders (e.g., ferromagnetic and ferroelectric) intrinsically coexist in the material; (ii) surface charging or carrier density modulation, which can modify the electronic band structure of ultra-thin metallic layers or semiconductors; (iii) magneto-ionics, which refers to the voltage-driven ion motion typically across layers in contact with liquid electrolytes or ionic conductors (e.g., Gd_2O_3 or HfO_2); (iv) electrochemical redox reactions; and (v) strain-mediated coupling in piezoelectric/magnetostrictive heterostructures. Each of these approaches faces some drawbacks: (i) dearth of available single-phase multiferroic materials and limited strength of magnetoelectric coupling at room temperature; (ii) limited penetration depth of electric field inside semiconductors and total electric-field screening at the surface of metals; (iii) low speed and limited endurance in magneto-ionic systems; (iv) lack of precise control of chemical reactions and their kinetics; and (v) clamping effects with the substrate and need of good interfaces in strain-mediated composite multiferroics. Despite these system-specific limitations, all the above mechanisms can be used to modify the properties of ferromagnetic materials (anisotropy, coercivity, saturation magnetization, Dzyaloshinskii-Moriya interaction, etc.), to a varying extent, with voltage.[5] Therefore, further progress is needed to address and overcome the current challenges. An added value to novel technologies is expected in piezoelectric/magnetostrictive heterostructures, where the induced direct magnetoelectric effects (that is, generation of electricity by applying magnetic fields) are appealing for energy harvesting/conversion applications and for wireless neuron/muscle cell stimulation. This example underscores the highly interdisciplinary character of magnetoelectric technologies, whose development has triggered a wealth of new research directions in physics, chemistry, electronics, and engineering, in general.

Recent advances in the understanding of magnetoelectric mechanisms and new materials with significant voltage-driven magnetic effects are reported in this Special Topic. State-of-the-art applications, including antennas, sensors, actuators, or magnetoelectric random-access memories, among others, are also described. Specifically, the Special Topic contains one Research Update, three Perspectives, and eight original research articles covering fundamental and application-oriented aspects of magnetoelectrics.

In the Research Update by Liang *et al.*,[6] the authors provide an extensive review of the existing magnetoelectric materials and devices, with emphasis on multiferroics, either single-phase or composites, both in bulk and thin-film form. The paper compares the magnetoelectric coupling strength for an extensive variety of materials and lists the values of magnetoelectric constants in the dynamic regime in a wide range of operational frequencies. The salient features of a range of magnetoelectric devices (antennas, sensors, random-access memories, energy harvesters, inductors, filters, etc.) are described, and the advantages with respect to other conventional systems not using magnetoelectric effects are emphasized.

In their Perspective, Nichterwitz *et al.*[7] put the focus on magneto-ionics, particularly on the achieved voltage coefficients for magnetization and coercivity and the demonstrated time scales for magneto-ionic switching. The authors overview the progress in oxygen magneto-ionics, using solid and liquid electrolytes, while the use of other ion species (nitrogen, hydrogen, etc.) is also mentioned. The challenges of using this type of materials for real applications (in terms of switching rates and endurance) are discussed.

Voltage-driven magneto-ionic control in ferroic heterostructures using ionic liquids and ionic conductors is also overviewed in the Perspective by Gu *et al.*[8] A particular aspect of this work is that it reports on the usefulness of electrostatic and electrochemical methods in tuning the ferroelectric properties of oxide-containing heterostructures. The potential of magneto-ionics in tuning the properties of skyrmions, van der Waals magnets, and topological Hall effects is also considered, as well as its applicability in healthcare technologies and synaptic devices (i.e., neuromorphics).

In the Perspective by Nicolenco *et al.*,[9] the advantageous effects of "strain gradients" (compared to homogeneous strains) in certain multiferroic heterostructures (such as vertically aligned composites or multi-phase porous frameworks) are reviewed. Although this kind of materials are still in their early years, the possibility to combine flexoelectricity with magnetostriction offers additional opportunities to the research community, such as the development of voltage-controlled functionally graded materials or new approaches for more efficient biomedical platforms.

Examples of scalable and cost-effective synthetic methods to prepare magnetoelectric heterostructures are reported in the original papers by Samghabadi *et al.*[10] and Mo *et al.*[11] Both of them use $CoFe_2O_4$ structures as a ferrimagnetic counterpart, coupled with $BaTiO_3$ and $BiFeO_3$, respectively. Interestingly, both converse coupling and direct magnetoelectric coupling is demonstrated in the $CoFe_2O_4/BiFeO_3$ system,[11] which consists of self-assembled $CoFe_2O_4$ slab-shaped crystals embedded in the $BiFeO_3$ matrix.

Electric-field-induced non-volatile magnetic switching in a $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) thin film is investigated by Wilhelm *et al.*[12] Detailed photoemission electron microscopy observations are used to correlate chemical with magnetic voltage-driven effects in this system. Rouco *et al.*[13] reported on the independent control of ferroelectric and oxygen vacancy switching in multiferroic tunnel junctions with the LSMO bottom electrode, $BaTiO_3$ ferroelectric barrier, and Ni top electrode (leading to a controlled inversion of the interface spin polarization), an effect that could open interesting new avenues toward neuromorphic devices. The use of novel ion species, such as hydrogen or fluorine, surpassing some of the characteristics of oxygen ion migration, is explored in the works by Goessler *et al.*[14] and Vasala *et al.*[15] The former studied the influence of hydrogen intercalation (using aqueous electrolytes) on the magnetic properties of nanoporous $Pd_{(1-x)}Co_x$, with Co being located in superparamagnetic clusters. Temperature-dependent magnetization curves show that interstitial hydrogen atoms lead to an increase in magnetic anisotropy energy and a concomitant blocking of their magnetic moments (i.e., tuning the superparamagnetic blocking temperature). In turn, electrochemical fluorination of La_2CuO_4, in solid state, offers the possibility of precisely adjusting hole doping and thus tuning of the superdiamagnetic properties (perfect diamagnetic behavior of superconductors) of this compound.

Finally, included in the Special Topic are two papers dealing with specific applications of magnetoelectric materials. Nguyen *et al.*[16] showed the performance of cantilever magnetoelectric $Pb(Zr,Ti)O_3$/Tb–Fe–Co resonators for magnetic sensing applications. In turn, Spetzler *et al.*[17] studied the influence of the piezoelectric material on the signal and noise of magnetoelectric magnetic field sensors.

In conclusion, our Special Topic covers diverse aspects of magnetoelectricity and highlights the recent discoveries in this compelling research area. We hope that the included manuscripts will provide new insights inspiring the development of new magnetoelectric materials. Moreover, this Special Topic is intended to serve as a forum for students and researchers to further explore new system designs and devices whose energy efficiency, performance, and functionalities will surpass the current state of the art.

REFERENCES

1. J. Puebla, J. Kim, K. Kondou, and Y. Otani, "Spintronic devices for energy-efficient data storage and energy harvesting," Commun. Mater. **1**, 24 (2020).
2. C. Song, B. Cui, F. Li, X. Zhou, and F. Pan, "Recent progress in voltage control of magnetism: Materials, mechanisms, and performance," Prog. Mater. Sci. **87**, 33 (2017).
3. C. Navarro-Senent, A. Quintana, E. Menéndez, E. Pellicer, and J. Sort, "Electrolyte-gated magnetoelectric actuation: Phenomenology, materials, mechanisms, and prospective applications," APL Mater. **7**, 030701 (2019).
4. A. Molinari, H. Hahn, and R. Kruk, "Voltage-Control of magnetism in all-solid-state and solid/liquid magnetoelectric composite," Adv. Mater. **31**, 1806662 (2019).
5. J.-M. Hu and C.-W. Nan, "Opportunities and challenges for magnetoelectric devices," APL Mater. **7**, 080905 (2019).
6. X. Liang, H. Chen, N. Xiang Sun, "Magnetoelectric materials and devices," APL Mater. (in press) (2021).

[7]M. Nichterwitz, S. Honnali, M. Kutuzau, S. Guo, J. Zehner, K. Nielsch, and K. Leistner, "Advances in magneto-ionic materials and perspectives for their application," APL Mater. **9**, 030903 (2021).
[8]Y. Gu, C. Song, Q. Wang, W. Hu, W. Liu, F. Pan, and Z. Zhang, "Emerging opportunities for voltage-driven magneto-ionic control in ferroic heterostructures," APL Mater. (in press) (2021).
[9]A. Nicolenco, M. de h-Óra, C. Yun, J. MacManus-Driscoll, and J. Sort, "Strain-gradient effects in nanoscale-engineered magnetoelectric materials," APL Mater. **9**, 020903 (2021).
[10]F. S. Samghabadi, L. Chang, M. Khodadadi, K. S. Martirosyan, and D. Litvinov, "Scalable, cost-efficient synthesis and properties optimization of magnetoelectric cobalt ferrite/barium titanate composites," APL Mater. **9**, 021104 (2021).
[11]Z. Mo, G. Tian, W. Yang, S. Ning, C. A. Ross, X. Gao, and J. Liu, "Magnetoelectric coupling in self-assembled $BiFeO_3$-$CoFe_2O_4$ nanocomposites on (110)-$LaAlO_3$ substrates," APL Mater. (in press) (2021).
[12]M. Wilhelm, M. Giesen, T. Duchoň, M. Moors, D. N. Mueller, J. Hackl, C. Baeumer, M. H. Hamed, L. Cao, H. Zhang, O. Petracic, M. Glöß, S. Cramm, S. Nemšák, C. Wiemann, R. Dittmann, C. M. Schneider, and M. Müller, "Photoemission electron microscopy of magneto-ionic effects in $La_{0.7}Sr_{0.3}MnO_3$," APL Mater. **8**, 111102 (2020).
[13]V. Rouco, F. Gallego, D. Hernandez-Martin, D. Sanchez-Manzano, J. Tornos, J. I. Beltran, M. Cabero, F. A. Cuellar Jimenez, D. Arias, G. Sanchez-Santolino, F. J. Mompean, M. Garcia-Hernandez, A. Rivera-Calzada, M. Varela, M. C. Muñoz, C. Leon, Z. Sefrioui, and J. Santamaria, "Ferroionic inversion of spin polarization in a spin memristor," APL Mater. **9**, 031110 (2021).
[14]M. Goessler, S. Topolovec, H. Krenn, and R. Wuerschum, "Nanoporous $Pd_{1-x}Co_x$ for hydrogen-intercalation magneto-ionics," APL Mater. **9**, 041101 (2021).
[15]S. Vasala, L. Alff, and O. Clemens, "Tuning of superconductivity and superdiamagnetism of La_2CuO_4 by solid-state electrochemical fluorination and defluorination," APL Mater. **9**, 041107 (2021).
[16]N. T. Nguyen, G. Agnus, S. Matzen, T. Maroutian, Do T. H. Giang, and P. Lecoeur, "Cantilever magnetoelectric PZT/Tb-Fe-Co resonators for magnetic sensing applications," APL Mater. **9**, 041103 (2021).
[17]B. Spetzler, J. Su, R.-M. Friedrich, F. Niekiel, S. Fichtner, F. Lofink, and F. Faupel, "Influence of the piezoelectric material on the signal and noise of magnetoelectric magnetic field sensors based on the delta-E effect," APL Mater. **9**, 031108 (2021).