## Comment on "Superspin Glass Mediated Giant Spontaneous Exchange Bias in a Nanocomposite of $BiFeO_3$-$Bi_2Fe_4O_9$"

The exchange anisotropy at ferromagnetic-antiferromagnetic interfaces reflects in exchange bias (EB). The latter is customarily manifested after an external magnetic field is applied either when the system is cooled trough the Néel temperature, at the time the interface is created or during some posterior treatment. Besides, it has been suggested that a so-called spontaneous EB may be set by the first magnetic field applied to certain systems [1]. In a recent Letter, Maity *et al.* [2] allege that such behavior was observed in a $BiFeO_3$-$Bi_2Fe_4O_9$ nanocomposite. In the present Comment, we argue that the authors have not provided convincing evidences for the claimed spontaneous EB, which could well be an artifact. We will limit our criticism to three crucial points, leaving apart important issues such as the validity of the key assumption in the criticized Letter for the ferromagnetic core in $Bi_2Fe_4O_9$ nanoparticles which is based on misquoting the literature data [3] (for the magnetic structure of the bulk material see Ref. [4]) or the reliability of the measured quantities, all of them provided without error bars.

First, none of the hysteresis loops of the samples for which EB is claimed has been measured using magnetic fields high enough to reach unambiguous reversible magnetization. Thus, there are no proofs that the loop shift along the field axis, $H_E$, is actually EB. Most probably, it is only a minor loop manifestation [5] which makes the commented Letter disputable.

Second, the observed $|H_E|$ depends on the direction of the magnetic field first applied during the hysteresis loop trace as well as on that used during the field cooling (FC). Yet, it is unclear whether the very same sample or different "virgin" ones have been used in consecutive measurements. Let consider the following situations. (i) If the same virgin sample has been measured starting from either positive or negative fields, with the loops minor and the demagnetizing protocol not ideal [6], the distinct $|H_E|$ values are naturally attributed to dissimilar initial magnetic states. Furthermore, sample A presents training, which could also add to the difference in $|H_E|$. (ii) If different samples were used, the notion of a positive or negative field is meaningless since, for an isotropic as-made nanocomposite, any direction of the first applied field is equivalent [7]; the same holds for the FC. (iii) If the same sample is field cooled twice, an interfacial configuration set during the first FC might not be completely reversed during the second one, leading to different values of $|H_E|$.

The third point we choose to discuss is the attribution of the asymmetry in $H_E$ to a ferromagnetic-antiferromagnetic coupling mediated by a superspin glass (SSG) interface, whose existence has been sustained by two arguments. One of them is the dependence of $H_E$ on the number of consecutively measured loops (training effect). The better fit obtained via Eq. (1) instead of the empirical law from Ref. [8] is used to support the statement. However, even the smallest field step employed is bigger than the difference between the two fitting curves in Fig. 2(b).

The other argument concerns the use of the stop-and-wait protocol [9], where the magnetic susceptibility, $\chi^{\mathrm{ref}}_{\mathrm{ZFC}}(T)$, obtained after zero-field cooling (ZFC) is compared with $\chi_{\mathrm{ZFC}}$, i.e., a curve traced after cooling the system to an intermediate temperature $T_i$, waiting for a given time at $T_i$ and then resuming the cooling process. A dip around $T_i$ in the $\chi_{\mathrm{ZFC}} - \chi^{\mathrm{ref}}_{\mathrm{ZFC}}$ vs $T$ curve is regarded as a SSG signature. In the work of Maity *et al.*, however, the process has been stopped during the reheating and not during the cooling as stated in [9], defining a completely different protocol. This might explain why a nonzero difference between the curves was observed for the entire displayed temperature interval and not only around the dip, as one should expect for SSG systems [9]. Thus, both evidences for the SSG phase are, at least, contestable.

Work supported by CNPq (projects 245674/2012-8, 307082/2012-1, 483277/2012-6, and 300961/2014-6).

A. Harres,[1,2] J. Geshev[1] and V. Skumryev[2]
[1]Instituto de Física-UFRGS
Porto Alegre 91501-970, RS, Brazil
[2]Institut Català de Recerca i Estudis Avançats (ICREA) and Departament de Física, Universitat Autònoma de Barcelona
08193 Barcelona, Spain

Received 19 December 2013; revised manuscript received 14 April 2014; published 4 March 2015
DOI: 10.1103/PhysRevLett.114.099703
PACS numbers: 75.70.Cn, 75.75.-c

[1] J. Saha and R. H. Victora, Phys. Rev. B **76**, 100405(R) (2007); B. M. Wang, Y. Liu, P. Ren, B. Xia, K. B. Ruan, J. B. Yi, J. Ding, X. G. Li, and L. Wang, Phys. Rev. Lett. **106**, 077203 (2011).
[2] T. Maity, S. Goswami, D. Bhattacharya, and S. Roy, Phys. Rev. Lett. **110**, 107201 (2013).
[3] References quoted in [2] clearly postulate an antiferromagnetic(core)-ferromagnetic(shell) structure, which is a logical assumption stemming from possible canting of magnetic moments at the surface.
[4] E. Ressouche, V. Simonet, B. Canals, M. Gospodinov, and V. Skumryev, Phys. Rev. Lett. **103**, 267204 (2009).
[5] L. Klein, Appl. Phys. Lett. **89**, 036101 (2006); J. Geshev, J. Magn. Magn. Mater. **320**, 600 (2008); J. Geshev, Appl. Phys. Lett. **93**, 176101 (2008).
[6] In the Supplemental Material to Ref. [2], Fig. 10 is supposed to demonstrate the effectiveness of the demagnetizing procedure performed, without arguments why. Actually, what one sees are some mysterious jumps on the FC curve at about 180 K, which were left without comment.
[7] J. Geshev, L. G. Pereira, and V. Skumryev, Phys. Rev. Lett. **100**, 039701 (2008).
[8] D. Paccard, C. Schlenker, O. Massenet, R. Montmory, and A. Yelon, Phys. Status Solidi **16**, 301 (1966).
[9] M. Sasaki, P. E. Jönsson, H. Takayama, and H. Mamiya, Phys. Rev. B **71**, 104405 (2005).

 