

Title	Maity et al. reply Maity et al. reply to the comment on "Superspin glass mediated giant spontaneous exchange bias in a nanocomposite of BiFeO3–Bi2Fe4O9", A. Harres, J. Geshev, and V. Skumryev, Physical Review Letters, 114, 099703 (2015)
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University College Cork, Ireland Coláiste na hOllscoile Corcaigh Maity et al. reply: In the preceding Comment, Harres et al. [1] expressed their concerns about some of the results reported in our Letter [2]. In this Reply, we take up and address their comments point by point. The first and the most important issue is whether the hysteresis loops are minor. A minor loop is nonsymmetrical and is expected to exhibit significant vertical asymmetry as well [3,4]. The loops reported in our work do not exhibit any vertical asymmetry. The magnetizations corresponding to the positive and negative maximum fields  $(H_m)$  are equal (Fig. 1) within the field and temperature limits of our study. Moreover, the magnetization is clearly found to be reversible (magnetizations for forward and reverse branches of the loop are merged around the maximum field applied) in the present case (Fig. 1). It is true that within the field limit applied, complete saturation of the magnetization could not be observed due to the presence of antiferromagnetic BiFeO<sub>3</sub> by a larger volume fraction. Incomplete saturation has been observed in a variety of systems where ferromagnetism is weak and associated with other magnetic phases, especially spin glass. The spin structure evolves here continuously with the field and the exchange bias is found to depend on the applied field [3,5]. Reference [6] contains the blown up portions of the hysteresis loops around the high field region. Hence, the doubt raised in the Comment is not well founded.

In response to the second point, we mention the following. The measurement of path dependency of the spontaneous exchange bias  $(H_E)$  has been carried out on the same sample. The sample has been appropriately demagnetized using the oscillating magnetic field with decreasing amplitude to ensure the identical initial magnetization state prior to each measurement. In fact, this issue has been rechecked by carrying out the measurements following zero field cooling from above 700 K (Supplemental Material in [2]; see also Ref. [6]) which is above the magnetic transition points. Therefore, the initial magnetization states of the sample did not differ in



FIG. 1 (color online). The magnetization values at the positive and negative maximum fields  $(H_m)$  for different  $H_m$ s; the extent of irreversibility as a function of magnetic fields for the loops measured at 5 and 300 K are also shown.

the measurements carried out to examine the dependence of  $H_E$  on the direction of the applied field. Since the path dependency has been clearly observed in the case of spontaneous exchange bias, its observation in the case of conventional exchange bias is indisputable.

The third point concerns the presence of the superspin glass phase. Although the memory effect has not been measured using the original protocol [7], the implication of which will be addressed separately, we point out that (i) we checked the fitting of the training curve by measuring the hysteresis loops with even smaller field step and found the validity of Eq. (1), (ii) the clear signature of the spin glass transition (frequency dependence of the transition temperature in ac susceptibility measurement) could be noticed in a similar system [8], and, finally and more importantly, (iii) the subtraction of the contribution of the paramagnetic component from the overall field-cooled magnetization yields a temperature independent pattern [7] at low temperature [Fig. 3(c) [2]] offering unambiguous evidence for the presence of the superspin glass phase in sample A. It is worth mentioning here that only sample A with a higher volume fraction of the superspin glass phase exhibits a large spontaneous exchange bias.

We further mention that a stronger ferromagnetic component is expected in even finer particles used in our study (~19 nm) than what has been used in the earlier work on nanoscale Bi<sub>2</sub>Fe<sub>4</sub>O<sub>9</sub> [9,10]. Finally, we point out that the error bar for the parameters such as  $H_F$ ,  $H_C$ , etc. is  $\pm 1\%$ -3%.

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