Observation of tunable exchange bias in Sr$_2$YbRuO$_6$

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The double perovskite compound, Sr$_2$YbRuO$_6$, displays reversal in the orientation of magnetic moments along with negative magnetization due to an underlying magnetic compensation phenomenon. The exchange bias (EB) field below the compensation temperature could be the usual negative or the positive depending on the initial cooling field. This EB attribute has the potential of getting tuned in a preselected manner, as the positive EB field is seen to crossover from positive to negative value above $T_{\text{comp}}$.

The notion of an exchange bias (EB) field$^1$, which is exemplified by the identification of a shift$^2$ in the centre of gravity of the magnetization hysteresis ($M$-$H$) loop, has implications for magnetic read heads$^3$, thermally assisted magnetic random access memories$^4$, and other spintronics devices$^5$$^6$. The EB phenomenon is considered to be a quintessential attribute$^4$ of a ferromagnetic (FM)/antiferromagnetic (AFM) composite or a bilayer assembly, when the entire system is cooled below ordering temperature $T_N$ of the AFM part, which lies below the ($T_C$) of the FM, i.e., $T_N < T_C$. Usually, the EB field is negative as the centre of $M$-$H$ loop gets left shifted below ($T_N$)$^1$. But occasionally the right shift of the loop, i.e., positive exchange bias (PEB) field has also been noted$^7$$^{12}$. The EB had mostly been observed in magnetic multi-component systems, such as oxidized magnetic nanoparticles, FM/AFM multilayers, FM/spin glass bilayer thin films, etc.$^1$. However interestingly, in recent years, it has also been noted in several bulk homogenous materials, like, manganites$^{13}$, cobaltates$^{14}$ and admixed intermetallic compounds$^{15}$$^{16}$. Recently Kulka$^{17}$ et al. have unearthed EB effect in a single crystal of an admixed rare earth intermetallic compound, Nd$_{0.75}$Ho$_{0.25}$Al$_2$, in close proximity to the compensation temperature ($T_{\text{comp}}$) in it. They encountered the sudden surfacing of the PEB on approaching $T_{\text{comp}}$, the EB changed sign on going across it; the sign reversal of exchange bias field is sought to be rationalized in terms of reversal in the nearly balanced local magnetic moment contributions from antiferromagnetically linked Nd$^{3+}$ and Ho$^{3+}$ ions along with that of the conduction electron polarization (CEP), with respect to the applied field direction$^{17}$.

We have now studied the EB phenomenon in a double perovskite antiferromagnetic (AFM) insulator Sr$_2$YbRuO$_6$$^{18}$. This compound belongs to the Ruthenates family, Sr$_2$LnRuO$_6$ ($Ln = Y$ or rare earth$^{19}$$^{21}$), and shows interesting magnetic properties below the magnetic ordering temperature, which include the (i) magnetization reversal and (ii) the magnetic compensation characterized by crossover of the magnetization axis ($M = 0$ line) towards the negative magnetization values$^{18}$. In the antiferromagnetic Sr$_2$YRuO$_6$ compound, containing non-magnetic Y element, the local moment resides on Ru ion alone$^{22}$, whereas in Sr$_2$YbRuO$_6$, both Yb and Ru ions possess local magnetic moments of 4.54 $\mu_B$ and 3.87 $\mu_B$, respectively$^{23}$. The monoclinic structure$^{23}$ of these AFM insulators facilitates the Dzyaloshinsky–Moria (D–M) interaction$^{24}$ between the antiferromagnetically ordered local moments. The anisotropic D–M interaction results in canting of the antiferromagnetically ordered moments and hence a weak residual ferromagnetism germinates$^{25}$$^{26}$. The presence of a weak ferromagnetic component could provide a circumstance analogous to that associated with the FM/AFM composite systems. Though the magnetization reversal, zero magnetization and negative magnetization in Sr$_2$YbRuO$_6$ could be expected from the combined effects of D–M interaction and the unidirectional anisotropy$^{18}$, the discovery of the EB and its sign reversal near the compensation temperature and the dependence of sign of EB at low temperature (i.e., below the $T_{\text{comp}}$) on the thermo-magnetic history has brought to light the possibility of tuning of the EB from positive to negative values in a convenient and predictable manner in double perovskite compounds. Such an interesting finding is being reported here; this attribute could be utilized in niche applications in spintronics.

A typical field-cooled (FC) magnetization curve $M_F$ measured at low fields (e.g. at \(H = 50 \text{ Oe}\)) in Sr$_2$YbRuO$_6$ is shown in Fig. 1. The sudden increase in the $M_F$ at the onset of the ordering temperature (44 K) is due to the presence of a weak ferromagnetic component$^{18}$ in this compound. The start of decrease in magnetization around $T \sim 39$ K is due to the onset of reversal in magnetization of the residual ferromagnetic part. This is triggered by the D–M interaction, as noted earlier in the case of isostructural Sr$_2$YRuO$_6$ compound$^{27}$. On cooling below 39 K, the decrease in magnetization continues and it crosses the $M = 0$ axis at the compensation temperature of $\sim 33.5$ K (marked as $T_{\text{comp}}$), and magnetization values move towards negative values on further lowering the temperature, signalling the importance of the role of Yb moments in Sr$_2$YbRuO$_6$; no crossover of $M = 0$ axis occurs in Sr$_2$YRuO$_6$, where Y$^{3+}$ ions are non-magnetic.

The two insets in Fig. 1 show the $M$-$H$ loops recorded between $\pm 50$ kOe at 5 K in Sr$_2$YbRuO$_6$ for two typical initial cooling fields, viz., 2 kOe and 50 kOe. At very high fields ($H > 20$ kOe), the $M$-$H$ response in both the insets of Fig. 1 are quasi-linear, as in antiferromagnets. However, at lower fields (i.e., between $\pm 20$ kOe), one can notice the presence of hysteresis bubbles, with small remnant magnetization ($0.05 \mu_B$/f.u. of Sr$_2$YbRuO$_6$), which can be attributed to the residual ferromagnetic component originating from a
slight canting of the antiferromagnetically coupled large moments of Yb and Ru ions. There is a subtle but important difference between the two bubbles in the two insets of Fig. 1, which pertains to the shift in the centre of gravity of these bubbles. Figure 2 shows the central portions of two hysteresis bubbles plotted together on an expanded scale, a noticeable difference between the dashed and solid curves as they cross the $M = 0$ axis at a magnetic field ($H_{-}$ or $H_{+}$) can be immediately recognized. The difference between the two loops in Fig. 2 can be characterized in terms of an exchange bias field ($H_E$), which is usually defined as $H_E = (H_{-} + H_{+})/2$, where $H_{-}$ and $H_{+}$ are the field values at which magnetization axis is crossed during the descending and the ascending field cycles. The loop measured between $\pm 50$ kOe in a cooling field ($H_{FC}$) of 2 kOe shows that the offset field $H_E$ is positive and the $M$-$H$ loops is slightly right shifted, whereas, in that with $H_{FC} = 50$ kOe the offset field, $H_E$, is negative. The two $H_E$ values (positive/negative exchange bias) with two different initial FC values are marked in Fig. 2.

To explore the effect of $H_{FC}$ on $H_E$, $M$-$H$ loops were later traced between $\pm 15$ kOe as a function of $H_{FC}$ at two temperatures, 5 K (i.e., well below the compensation temperature $T_{comp}$ of 33.5 K) and 35 K ($> T_{comp}$) as a function of the cycling field ($H_{FC}$). The $H_E$ values obtained from such measurements are shown in Figs. 3(a) and 3(b). At 5 K (cf. Fig 3(a)), $H_E$ is positive for low cooling fields. As the value of $H_{FC}$ increases, $H_E$ monotonically decreases, crossing zero at $H_{FC} = 10$ kOe, and thereafter moves towards a saturated negative value, above $H_{FC}$ of 30 kOe. On the other hand, at $T = 35$ K, one observes only the usual negative $H_E$ values in low as well as high cooling fields (cf. Fig 3(b)). We also noted that a given $H_E$ value depends on the extent of the limit set on the cyclic field (e.g. $\pm 15$ kOe for data in Fig. 3(a)), however the trend of change in sign of $H_E$ as a function of $H_{FC}$ (at 5 K) and its approach to saturation at $T_{comp}$ at 35 K does not depend on the choice of the limit set on the cycling field.

The temperature variation study of the EB effect was performed on the hysteresis loops measured between $\pm 15$ kOe in two cooling fields, 50 kOe and 2 kOe. $H_E$ values obtained from the hysteresis loops at different temperatures are shown in Fig. 4(a) along with the half width of the hysteresis loops, which is often termed as the effective coercive field, $H_E^{eff} = -(H_{-} - H_{+})/2$, in Fig 4(b) for comparison. We note first in Fig. 4(b) that just as in the two insets of Fig. 1 for 5 K, the width of hysteresis loop does not imbibe any dependence on the initial cooling field values at all temperatures. $H_E^{eff}(T)$ in Fig. 4(b) displays a minimum at about 37 K (see, also, inset of Fig. 4(b)), which is unexpectedly above the compensation temperature of 33 K in low fields (cf. Fig. 1)\textsuperscript{18}. $T_{comp}$ values had earlier been noted\textsuperscript{18} to decrease with large enhancement in $H$ ($H > 2$ kOe) in this system. As regards the temperature dependence of EB field in Fig. 4(a), the $H_E$ values for $H_{FC} = 50$ kOe maintained the usual negative sign over entire temperature range on warming up from 5 K towards $T_C$. However, those for $H_{FC} = 2$ kOe show a sign reversal from positive to

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**Figure 1:** Temperature variation of the field-cooled (FC) magnetization in $H = 50$ Oe in Sr$_2$YbRuO$_6$. The $T_C$ and $T_{comp}$ values have been identified. The insets (a) and (b) show the magnetic hysteresis ($M$-$H$) loops traced at 5 K after initial field cooling in (a) $H_{FC} = 2$ kOe and (b) 50 kOe, respectively.

**Figure 2:** Expanded portion of the magnetization loops in cooling fields ($H_{FC}$) of 2 kOe and 50 kOe in Sr$_2$YbRuO$_6$ at 5 K. Note that the exchange bias field $H_E$ in the two cases have opposite signs.

**Figure 3:** Dependence of the exchange bias field ($H_E$) on the initial cooling field ($H_{FC}$) in Sr$_2$YbRuO$_6$ at (a) 5 K and (b) 35 K.
positive values at about 32 K, which is close to the $T_{\text{comp}}$ value in Fig. 1. The latter behaviour appears to echo the change in sign of $H_E$ reported across $T_{\text{comp}}$ in the admixed rare earth intermetallic Nd$_{0.75}$Ho$_{0.25}$Al$_2$ and other similar systems which imbibe compensation behaviour. However, the sense of sign change in Sr$_2$YbRuO$_6$ is phase reversed vis. a vis. that in Nd$_{0.75}$Ho$_{0.25}$Al$_2$. The $|H_E|$ values for $H_{\text{FC}} = 50$ kOe reach a local minimum near $T_{\text{comp}}$ (cf. Fig. 4(a)). Above about 35 K, the two sets of $H_E(T)$ values (in Fig. 4(a)) overlap, thereby implying their independence of the initial field cool history. The $|H_E(T)|$ values can be seen to reach a local maximum at 37 K, where $H_E^{\text{eff}}(T)$ is minimum. This correspondence between $H_E^{\text{eff}}(T)$ and $|H_E(T)|$ is also very different from the trend seen in the admixed rare-earth intermetallic and the magnetic multilayer systems across the respective $T_{\text{comp}}$ values. In the latter varieties of systems, the width of the hysteresis loop nearly collapses at $T_{\text{comp}}$, and the exchange bias also tends to disappear.

In consonance with the vanishing of the magnetization at the compensation temperature, Fig. 4(a) also shows that while cooling down $|H_E(T)|$ surfaces up only on cooling field below 41 K, the temperature at which $H_{\text{comp}}^{\text{eff}}(T)$ shows a local maximum. It has been argued earlier that the decrease in magnetization below 41 K signals the onset of turnaround in the orientation of local moments of Ru due to coming in the play of the D–M interaction. We believe that subtle differences between the details of correspondence between the $H_E^{\text{eff}}(T)$ and $H_E(T)$ occurring in Sr$_2$YbRuO$_6$ vis. a vis. other systems, where compensation behavior has been noted, are a consequence of competition and interplay between three components in Sr$_2$YbRuO$_6$ (viz. (i) residual ferromagnetism from Ru and (ii) residual magnetisation from Yb moments, and (iii) reorientation in Ru and Yb moments triggered by the D–M interaction) instead of the usual competition between the two antiferromagnetically linked components in other systems.

To summarize, we have shown that an antiferromagnetic double perovskite compound Sr$_2$YbRuO$_6$ comprising magnetic moments residing on rare earth and transition elements exhibits a large variety in exchange bias behaviour. The dependence of EB field on the cycling field, its sign change on moving across $T_{\text{comp}}$, dependence of EB on the cooling field, etc. are attributes, which have been noted earlier in different magnetic multi-components systems, in the forms of multilayer, coated nano particles, etc. as well as in single component systems, like, admixed rare earth intermetallics, transition metal oxides, etc. Our above findings that an antiferromagnetic double perovskite compound Sr$_2$YbRuO$_6$, exhibits the entire spectrum of exchange bias behaviour in the same compound, underscores the importance and novelty of oxide compounds comprising rare earth and transition metal ions. The exchange bias field of either sign can be selected in the Sr$_2$YbRuO$_6$ compound in a predetermined manner by choosing the cooling field and/or changing the temperature. The attribute of easy tuning of the exchange bias calls for further explorations of this behaviour in single crystal and/or oriented thin film form to gain deeper insight into the microscopic basis of the origin of exchange bias and its variation with field/temperature and thermomagnetic history in the compounds crystallizing in double perovskite structure.

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1. J. Nogues and I. K. Schuller, J. Magn. Magn. Mater. 192, 203 (1999).
2. W. H. Meiklejohn and C. P. Bean, Phys. Rev. 102, 1413 (1956).
3. C. Tsang, R. E. Fontana, T. Lin, D. Heim, V. S. Speriosu, B. A. Gurney, and M. L. Williams, IEEE Trans. Magn. 30, 3801 (1994).
4. I. L. Prejbeanu, M. Kerekes, R. C. Sousa, H. Sibuet, O. Redon, B. Dieny, and J. P. Nozières, J. Phys.: Condens. Matter 19, 165218 (2007).
5. Y. T. Chen, S. U. Jen, Y. D. Yao, J. M. Wu, J. H. Liao and T. B. Wu, J. Alloys Compounds 448, 59 (2008).
6. H. Fuke, Y. Kamiguchi, S. Hashimoto, T. Funiyama, K. Saito, H. Iwasaki and M. Sahashi, US Patent No 6057049 (2004).
7. J. Nogués, D. Lederman, T. J. Moran and I. K. Schuller, Phys. Rev. Lett. 76, 4624 (1996).
8. C. Leighton, J. Nogués, B. J. Jönsson-Akerman and I. K. Schuller, Phys. Rev. Lett. 84, 3466 (2000).
9. T. Gregid, I. N. Krivorotov, P. Eames and E. D. Dahlberg, Appl. Phys. Lett. 81, 1270 (2002).
10. F. Radu, M. Etzkorn, R. Siebrecht, T. Schmitte, K. Westerholt and H. Zabel, Phys. Rev. B 67, 134409 (2003).
11 M. Ali, P. Adie, C. H. Marrows, D. Greig, B. J. Hickey and R. L. Stamps, Nature Mater. 6, 70 (2007).
12 J. T. Kohlhepp, H. Wieldraaijer and W. J. M de Jonge, J. Mater. Res. 22, 569 (2007).
13 S. Karmakar, S. Taran, E. Bose and B. K. Chaudhuri, Phys. Rev B 77, 144409 (2008).
14 Y.-k. Tang, Y. Sun and Z.-h. Cheng, Phys. Rev B 73, 174419 (2006).
15 X. H. Chen, K. Q. Wang, P. H. Hor, Y. Y. Xue and C. W. Chu, Phys. Rev B 72, 054436 (2005).
16 P. D. Kulkarni, S. Venkatesh, A. Thamizhavel, V. C. Rakhecha, S. Ramakrishnan and A. K. Grover, IEEE Trans. 45, 2902 (2009).
17 P. D. Kulkarni, A. Thamizhavel, V. C. Rakhecha, A. K. Nigam, P. L. Paulose, S. Ramakrishnan and A. K. Grover, Euro Phys. Lett. 86, 47003 (2009).
18 R. P. Singh and C. V. Tomy, J. Phys.: Condensed Matter 20, 23520 (2008).
19 R. P. Singh, S. Marik, C. V. Tomy and A. K. Grover, Proc. Annual DAE SSP Symposium, 54, 1085 (2009).
20 R. P. Singh, Ph.D. Thesis, Indian Institute of Technology Bombay, Mumbai, India, 2010.
21 P. C. Donohue and E. L. McCann, Mater. Res. Bull. 12, 519 (1977).
22 P. D. Battle and W. J. Macklin, J. of Solid State Chem. 52, 138 (1984).
23 Y. Doi and Y. Hinatsu, J. Phys.: Condens. Matter 11, 4813 (1999).
24 L. D. Tung, M. R. Lee, G. Balakrishnan, and D. McK. Paul, Phys. Rev. B 75, 104404 (2007).
25 I. Dzyaloshinsky, J. Phys. Chem. Solids 4, 241 (1958).
26 T. Moriya, Phys. Rev. 120, 91 (1960).
27 R. P. Singh and C. V. Tomy, Phys. Rev. B 78, 024432 (2008).
28 D. J. Webb, A. F. Marshall, Z. Sun, T. H. Geballe and R. M. White, IEEE Trans. Magn. 24, 588 (1988).
29 Y. Ren, T. T. M. Palstra, D. I. Khomskii, E. Pellegrin, A. A. Nu- groho, A. A. Menovsky and G. A. Sawatzky, Nature 396, 441 (1998).