

Fig. S1 Training of the giant spontaneous exchange bias at room temperature along [001]. There is slight change of the exchange bias field after training. Exchange bias field after training is about -575 Oe, still larger than the one reported in BiFeO<sub>3</sub>-Bi<sub>2</sub>Fe<sub>4</sub>O<sub>9</sub> nanocomposites.

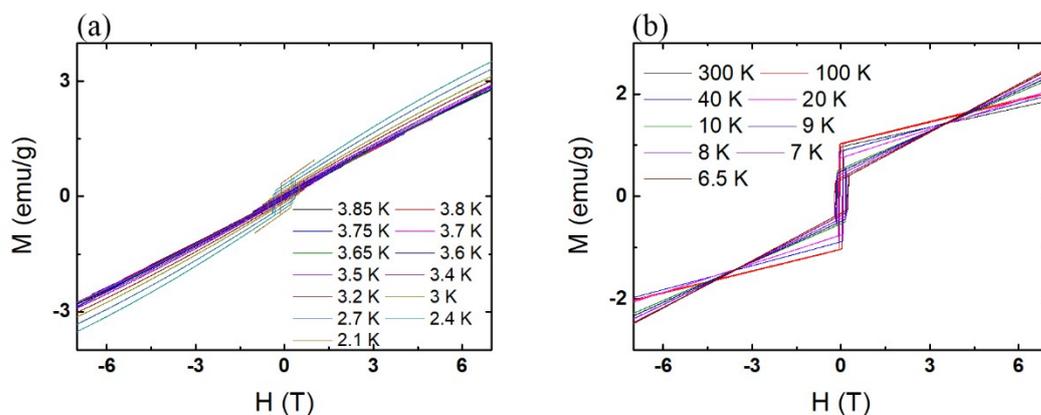


Fig. S2 Magnetic hysteresis loops (a) below 3.9 K and (b) above 6 K. Fe<sup>3+</sup> spin dominates above 3.9 K, so the slope becomes larger at lower temperature, while the remnant magnetization drops at lower temperature, which introduces crossing over the M-H lines at high magnetic field. While below 3.9 K, the Sm<sup>3+</sup> spin dominates, so the slope and remnant magnetization increase simultaneously at lower temperature, and no crossing can be notice then.

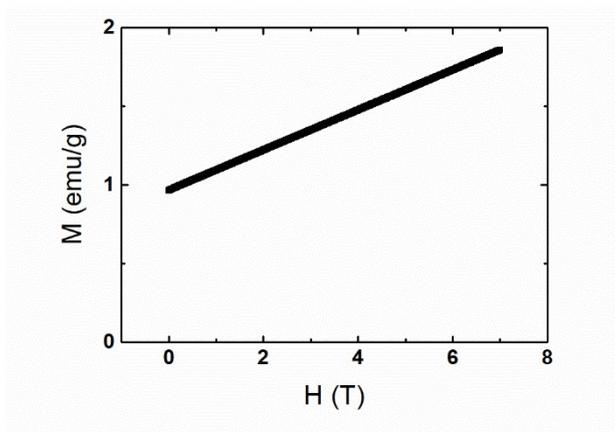


Fig. S3 Magnetization procedure under high magnetic field at room temperature. An obvious sloop can be seen. The hysteresis part is not shown, which is more clearly seen in Fig. 2 (a).

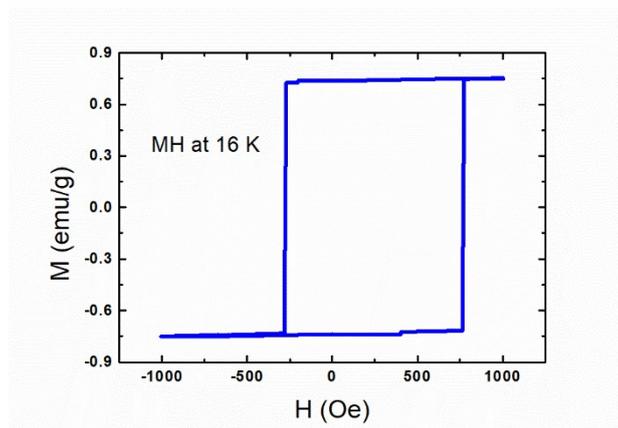


Fig. S4 ZFC Hysteresis loop at 16 K cooled from 750 K without any magnetic field. The possible residual magnetic field of the magnet is limited to several oersted, which is far below such exchange bias strength ( $\sim 250$  Oe).

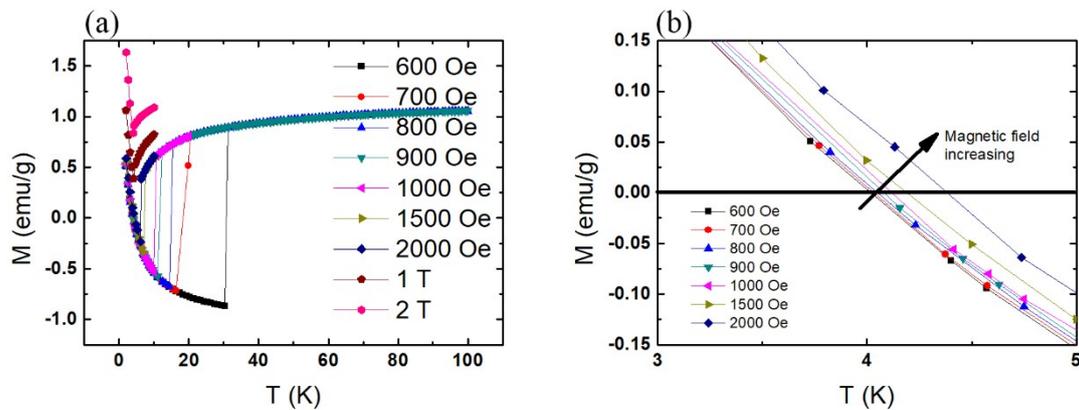


Fig. S5 M-T curves under different magnetic fields: (a) the jump of magnetization and (b) a detailed tendency of compensation point with variation of magnetic field. It can be seen that the compensation point does vary with magnetic field, which causes a little variation of the compensation point in different reports. However, the most appropriate value should be obtained under low magnetic field, at temperature of which the remnant magnetization becomes zero.

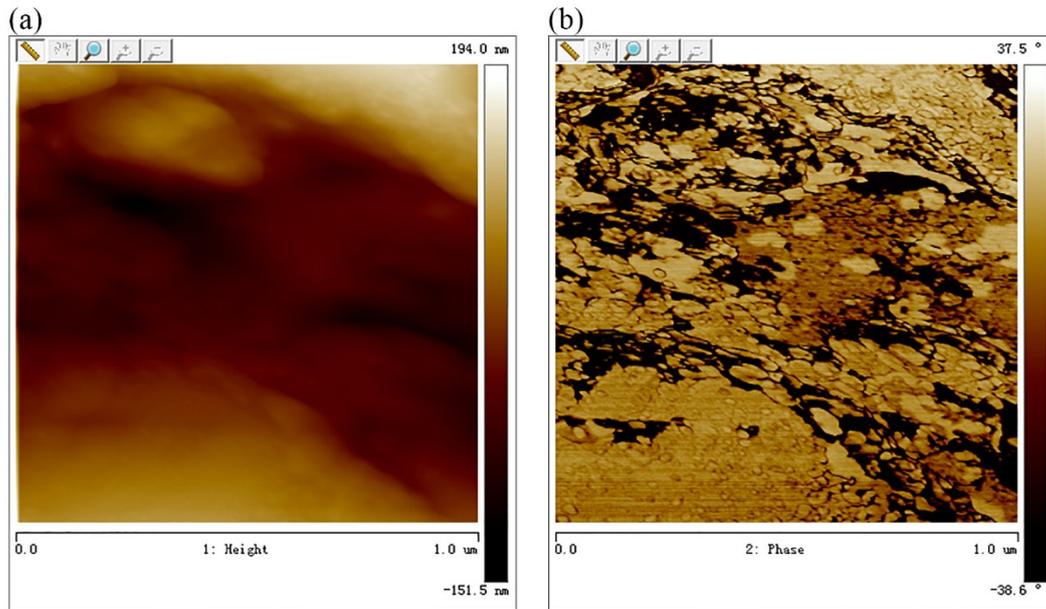


Fig. S6 Magnetic force microscopy (MFM) image of SFO. (a) Surface topography and (b) MFM phase contrast of the single crystal sample. It can be clearly seen that some magnetic phase separation does happen. This may originate from the crystal domain structure of the sample.

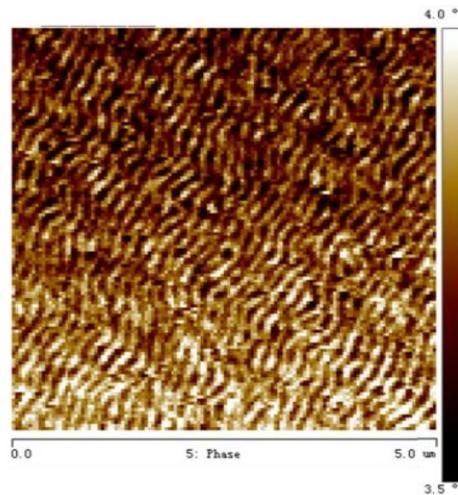


Fig. S7 MFM magnetic phase diagram of 50 nm thick Ni polycrystalline thin film.