

Supplementary Information

Nonascale structural modulation and the enhanced room-temperature multiferroic properties

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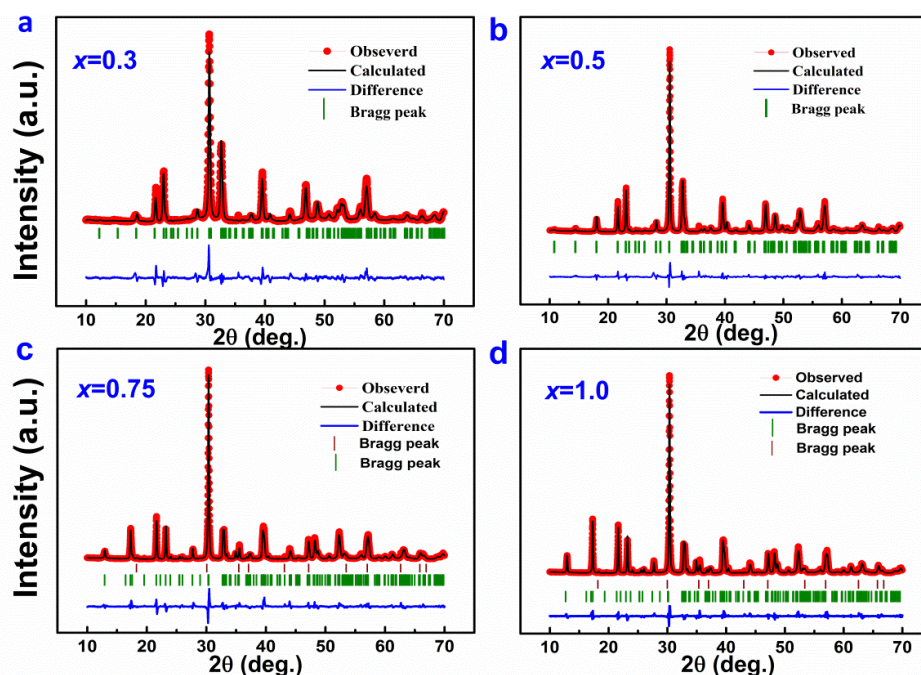
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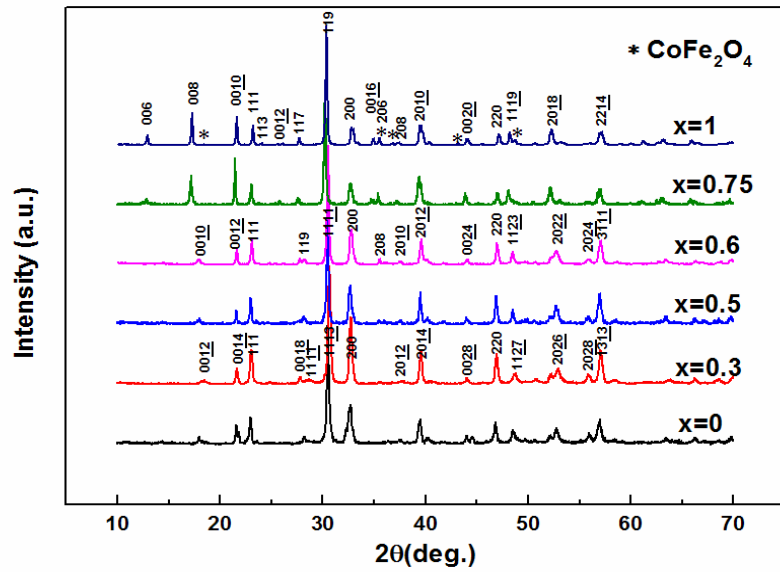
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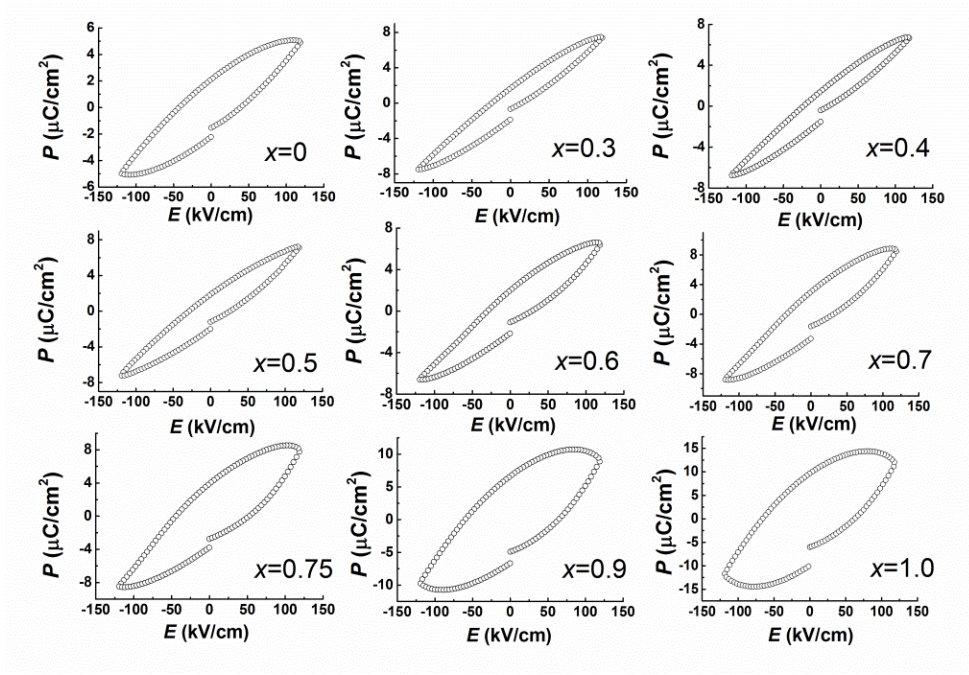
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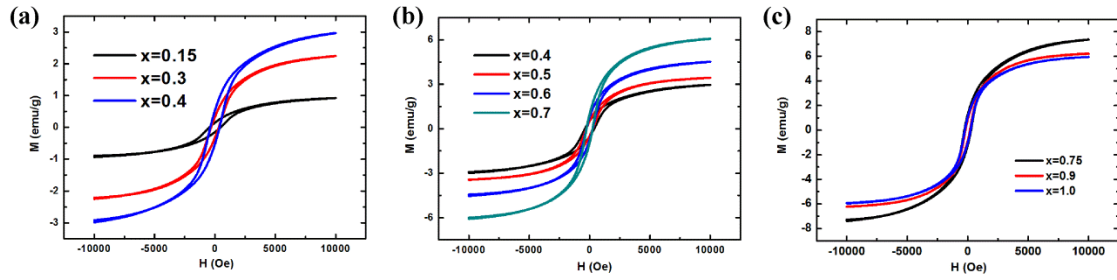
Supplementary Fig. S1 Rietveld refinements for the powder XRD patterns of the sintered BFCT-*x* ceramics. (a) BFCT-0.3, (b) BFCT-0.5, (c) BFCT-0.75, and (d) BFCT-1.0. These XRD patterns were refined in the orthorhombic lattice using GSAS software. Circles indicate the experimental data and calculated data are the continuous black line overlapping them. The blue curve shows the difference between the experimental and calculated patterns. The vertical bars indicate the expected reflection positions. The lattice parameters and space groups turn out to be $a=5.478$ Å, $b=5.488$ Å, $c=57.405$ Å and $Fm2m$ for $x=0.3$ ($R_p=11.8\%$), $a=5.458$ Å, $b=5.460$ Å, $c=49.257$ Å and $F2mm$ for $x=0.5$ ($R_p=10.3\%$), $a=5.462$ Å, $b=5.434$ Å, $c=41.115$ Å and $A2_1am$ for $x=0.75$ ($R_p=12.6\%$) and $a=5.465$ Å, $b=5.433$ Å, $c=41.113$ Å and $A2_1am$ for $x=1$ ($R_p=8.8\%$), respectively. Minor $CoFe_2O_4$ phase with space group $Fd-3m$ and $a=b=c=8.382$ Å is found in the samples of BFCT-0.75 and BFCT-1.0, in which the amount is below the XRD instrumental resolution.



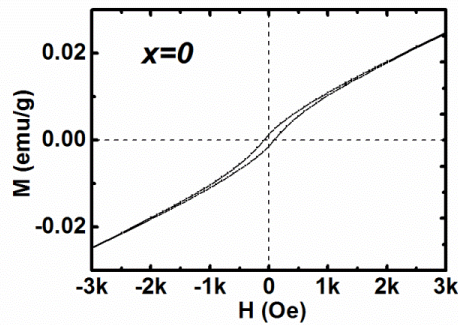
Supplementary Fig. S2 X-ray diffraction patterns of the sintered BFCT- x ($x=0, 0.3, 0.5, 0.6, 0.75, 1.0$) at room temperature. The peaks of $x=0$ and $x=0.3$ are indexed in accordance with $\text{Bi}_7\text{Fe}_3\text{Ti}_3\text{O}_{21}$ (JCPDS, No.54-1044), the peaks of $x=0.5$ and $x=0.6$ are confirmed from $\text{Bi}_5\text{Fe}_2\text{Ti}_3\text{O}_{21}$ (the space group $F2mm$), and the peaks of $x=0.75$ and $x=1.0$ are coincided with $\text{Bi}_5\text{FeTi}_3\text{O}_{15}$ (JCPDS, No.82-0063). The samples are all mainly with the Aurivillius phase, though the samples of $x=0.75$ and $x=1.0$ show minor CoFe_2O_4 impurity phase (marked by “*”).



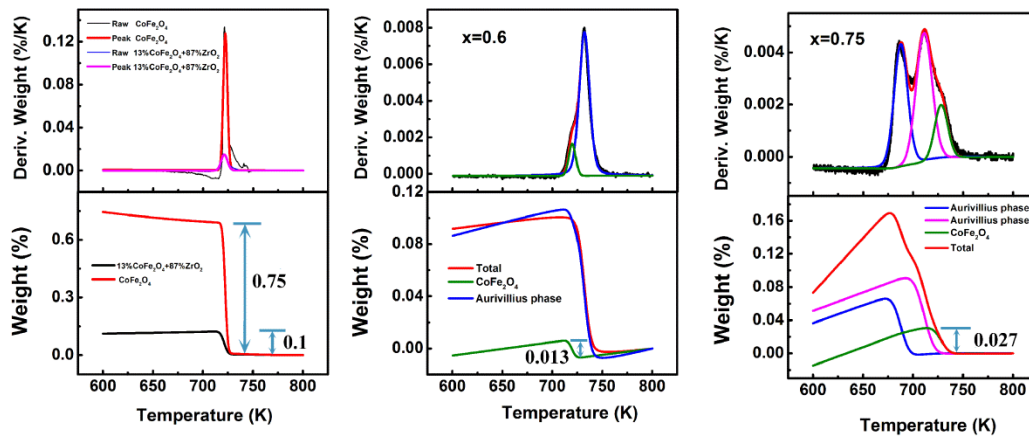
Supplementary Fig. S3 P - E loops of BFCT- x ceramics at the room temperature.



Supplementary Fig. S4 Hysteresis loops of magnetization (M) in the BFCT- x ($x \neq 0$) ceramics at the room temperature.



Supplementary Fig. S5 Magnetic hysteresis loops of BFCT-0 ceramic at the room temperature. $2M_r = 2.67$ memu/g.



Supplementary Fig. S6 Fitted DMTG curves and integral curves for CoFe_2O_4 , ZrO_2 -13wt% CoFe_2O_4 and BFCT- x ($x = 0.6$ and 0.75) at the temperature 600~800 K. The weight loss of CoFe_2O_4 is 0.75%, and the weight loss of ZrO_2 -13wt% CoFe_2O_4 is about 0.1%. Intriguingly, $0.1\% / 0.75\% = 13\%$, is consisted with the weight percentage of CoFe_2O_4 in the predesigned ZrO_2 -13wt% CoFe_2O_4 . In the terms of our analogy, the weight percentage content of CoFe_2O_4 in BFCT-0.6 and BFCT-0.75 is $\sim 1.7(\pm 0.5)\text{wt}\%$ ($0.013\%/0.75\%$) and $\sim 3.6(\pm 0.5)\text{wt}\%$ ($0.027\%/0.75\%$), respectively. By this method, the content of CoFe_2O_4 in other samples can be estimated ($\sim 0.5\text{wt}\%$ in BFCT-0.4, $\sim 1.0(\pm 0.5)\text{wt}\%$ in BFCT-0.5, $\sim 2.6(\pm 0.5)\text{wt}\%$ in BFCT-0.7 and $\sim 3.6(\pm 0.5)\text{wt}\%$ in BFCT-0.9~1.0).