

Self-biased magnetoelectric coupling in nano-microscale multiferroic composite $0.3\text{NiFe}_2\text{O}_4/0.7[0.948(\text{K}_{0.5}\text{Na}_{0.5})\text{NbO}_3-0.052\text{LiSbO}_3]$

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We examined the dependence of magnetoelectric (ME) voltage coefficient (α) on DC bias magnetic field in particulate $0.3\text{NiFe}_2\text{O}_4/0.7[0.948(\text{K}_{0.5}\text{Na}_{0.5})\text{NbO}_3-0.052\text{LiSbO}_3]$. The ME hysteresis loop was observed and the ratio of remanent α to maximum α is about 1: 2. The results also show that the values of remanent α and coercive fields of α are remain the same at different H ranges, and the giant self-bias phenomenon is relative to magnetic hysteresis of the materials.

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1. Introduction

Since the magnetoelectric effect was firstly pointed out by Curie in 1894 [1], various ME material systems and processing techniques have been developed to improve the ME effect properties for applications [2]. It has been conformed both experimentally and theoretically that the ME coupling in magnetostrictive-piezoelectric composites is much larger than that in single phase materials [3,4]. Recently, giant ME effect was practically achievable in a lot of ME composite materials such as $\text{Ba}_{0.5}\text{Sr}_{1.5}\text{Zn}_2(\text{Fe}_{1-x}\text{Al}_x)_{12}\text{O}_{22}$, $\text{Pb}_x\text{Sr}_{1-x}\text{TiO}_3$ and $\text{Pb}_n\text{Nb}_2\text{O}_{5+n}$ [5-7]. These findings provide a potential use in ME applications such as sensors, transducers, filters and so on [8-10].

Recent studies have revealed that a large α value can be obtained in the absence of DC bias magnetic field (H), so called self-biased α effect in some laminate composites [11-13]. These findings are encouraging this kind of materials for applications such as self-biased magnetic field sensors [13]. For ME composites, the ME effect is determined by the piezoelectric and piezomagnetic coefficients of the materials. The relationship between ME effect coefficient and piezomagnetic coefficient can be expressed as $\alpha \propto q = d\lambda/dH$, here α is the ME effect coefficient, q is piezomagnetic coefficients and λ is magnetostriction [14]. In addition, it is also confirmed that the ME effect coefficient is associated with the magnetic hysteresis loops in layered ME composite [14]. However,

up to now, the most studies on self-biased effect focused on the layered or laminate ME composites, while seldom report has been made on the particulate composites. Moreover, the relations between α and magnetic property is still lack deep investigation in particulate ME composite. In this paper, we present a study on self-biased ME effect of ME particulate composites $0.3\text{NiFe}_2\text{O}_4/0.7[0.948(\text{K}_{0.5}\text{Na}_{0.5})\text{NbO}_3-0.052\text{LiSbO}_3]$ (NFO/KNN), which consisting of ferroelectric and ferromagnetic materials. The dependence of ME effect on H_{bias} was investigated and the relations between α and magnetic property were also studied. It is expect that this research would bring more understanding and give some useful information on particulate ME composites.

2. Experimental details

The composites $0.3\text{NiFe}_2\text{O}_4/0.7[0.948(\text{K}_{0.5}\text{Na}_{0.5})\text{NbO}_3-0.052\text{LiSbO}_3]$ (NFO/KNN) were prepared. KNN was prepared by a solid-state reaction method, and NFO was synthesized using citrate gel method. The particulate composites NFO/KNN were prepared by mixing 30mol% of NFO phase with 70 mol% of KNN phase. The composite mixtures were pressed into pellets (thickness 1–2mm and 10mm diameter) using a hydraulic press and sintered at 1050°C for 4 h to yield the final products [15]. The particle morphological features were imagined by scanning electron microscopy (SEM, SU8010 FE-SEM).

For ME characterization, the samples were deposited on silver electrodes and then poled in silicone oil at a poling electric field from 8 to 13 kV/cm with poling temperature from 405 K to room temperature. The ME effect measurement was performed as described in Refs.16 and 17. The ME coefficient $\alpha=dE/dH$ was determined by the electric field induced under a small applied AC magnetic field superimposed onto the DC magnetic bias field [17]. Both the AC magnetic field and magnetic bias were parallel to the transverse direction of the samples. The magnetic property of the sample was detected by using vibrating sample magnetometer (VSM option on Versalab). All measurements were performed at room temperature.

3. Results and discussion

Fig. 1 shows the SEM micrograph and EDS spectrum of NFO/KNN. The ceramic shows a homogeneous microstructure. It seems that the NFO particles are distributed within KNN-LS matrix. From the small NFO particle in spectrum 1 and the big KNN-LS particle in spectrum 2, the coexistence of NFO and KNN phases in the composite is further evident. The particle size of NFO is less than 200nm, while that of KNN-LS is bigger than 10 μ m as shown in Fig. 1. The nano-microscale structure is confirmed.

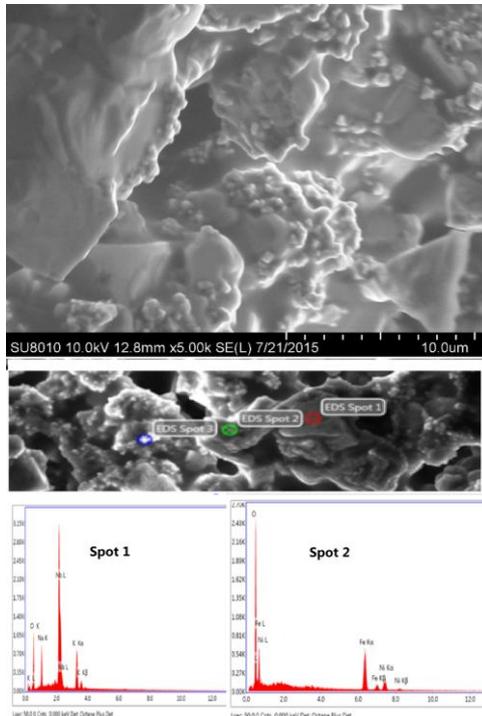


Fig. 1. SEM micrographs and EDS spectrum of NFO/KNN ceramic at room temperature. The coexistence of NFO and KNN phases in the ceramics is further evident by the big KNN particle in Spot 1 and the small KNN particle in Spot 2

Fig. 2(a) shows the change in ME voltage coefficients as a function of DC bias magnetic field (H) at frequency of 1 kHz. The α and phase vs H were measured by varying H from 1500 Oe to -1500 Oe, after that from -1500 Oe to 1500 Oe. As H is decreased from 1000 Oe to -1000 Oe, α initially increases to a positive peak value and then decreased to a negative peak value. As H is increased from -1000 Oe to 1000 Oe, α does not reach the previous value, exhibiting hysteresis behavior. The α shows a hysteretic behavior during H sweep with remanent α value of $\sim\pm 1.68$ mV \cdot cm $^{-1}$ \cdot Oe $^{-1}$ at $H=0$ as shown in Fig. 2(a). In addition, one can observe that the largest negative peak α of -3.23 mV \cdot cm $^{-1}$ \cdot Oe $^{-1}$ exactly appears at $H=250$ Oe, and the positive peak α of 3.35 mV \cdot cm $^{-1}$ \cdot Oe $^{-1}$ appears at $H=250$ Oe. The ratio of remanent α to maximum α is about 1: 2. It is a much big value for this kind of ME materials. The ME voltage coefficient with DC bias field exhibits hysteresis, and forming a loop in a test period. From the loop, the coercive field of α (H_c) were obtained on the basis of expression $H_c = (H_{c+} + |H_{c-}|)/2$, and the value is about 76.5 Oe. Fig. 2(b) shows the phasic difference as functions of H at the frequency of 1 kHz. When H is reversed, the phasic difference become opposite, which means that this kind of ME materials can be used to make phase transformer.

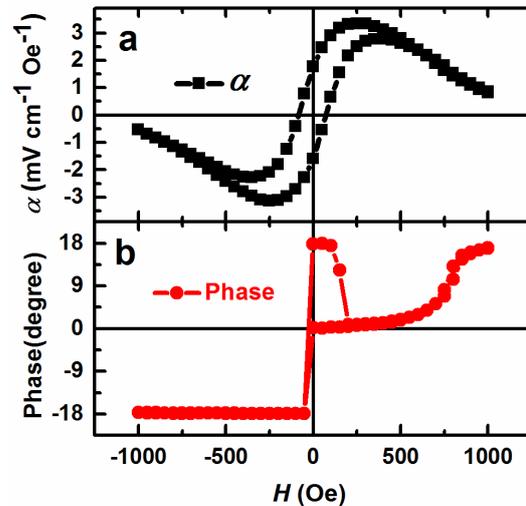


Fig. 2. Variations in transverse ME voltage coefficients and phasic difference with the bias magnetic field H for NFO/KNN at frequency of 1 kHz. The lines are guide to the eye.

Fig. 3 shows the H -dependent α at different H ranges from $-H_{max}$ to H_{max} for NFO/KNN. With the increase of the magnitude of H_{max} , both $\alpha_{//}$ and α_{\perp} show different loop shapes. However, the values of remanent α and coercive fields of α are almost unchanged. The result can be provided a reference for practical use. At $H_{max}=500$ Oe, the loop shape of α is similar to magnetic hysteresis loop, which means that the ME composite with ME hysteresis behavior have more potential applications.

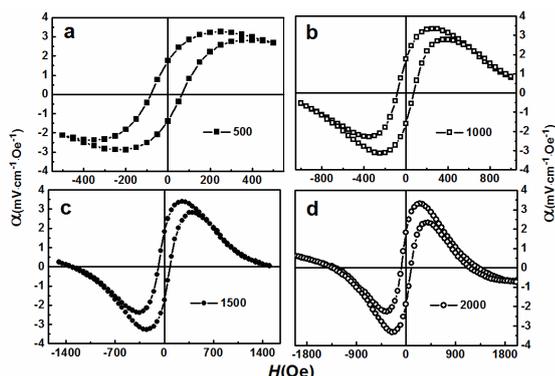


Fig. 3. H -dependent ME voltage coefficients at different DC bias magnetic ranges

Previous studies show that ME voltage coefficients is correlated with λ and M as $\alpha \propto d\lambda/dH \propto dM^2/dH$ [14]. The magnetization (M) as a function of magnetic field was shown in Fig. 4(a). To further confirm the relation $\alpha \propto dM^2/dH$, M^2 - H and (dM^2/dH) - H curves have been plotted as shown in Fig 4(b) and 4(c). It is observed that the behavior of dM^2/dH is similar to that of α . The data in Fig. 4 verify that the self-bias effect is mainly related to the magnetic hysteretic response of ferrite phase. This means that one can predict the self-bias ME effects by means of testing M - H loops. To get a high self-bias ME effect, one should choose the ME materials with a high quality magnetic properties.

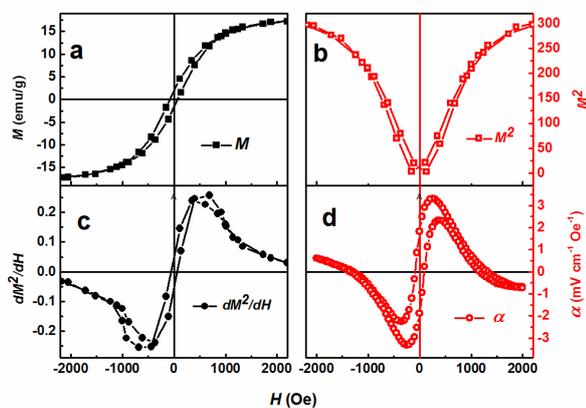


Fig. 4. (a) Magnetization-magnetic field (M - H) loops, (b) square of magnetization as a function of magnetic field, (c) differential of square magnetization-magnetic field $[(dM^2/dH)$ - H] for NFO/KNN. (d) DC bias magnetic field dependence of ME voltage coefficients

4. Conclusions

In summary, the dependence of DC bias magnetic field on ME voltage coefficient has been studied for nano-microscale multiferroic composites. The ME hysteresis loop was observed and the ratio of remanent α to maximum α is about 1: 2. It is also found that the values of remanent α and coercive fields of α are remain the same

at different H ranges. In addition, the composite exhibited giant self-bias phenomenon which was relative to magnetic hysteresis of the materials. These findings show that the nano-microscale multiferroic composite is a good candidate for potential application in ME multifunctional devices.

Acknowledgements

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References

- [1] P. Curie, J. Phys. Theor. Appl., **3**(1), 393 (1894).
- [2] M. Fiebig, Journal of Physics D: Applied Physics, **38**(8), R123 (2005).
- [3] C. W. Nan, M. I. Bichurin, S. Dong, D. Viehland, G. Srinivasan, Journal of Applied Physics, **103**(3), 031101 (2008).
- [4] Y. Yan, Y. Zhou, S. Priya, Applied Physics Letters, **102**(5), 052907 (2013).
- [5] W. S. Noh, K. T. Ko, S. H. Chun, K. H. Kim, B. G. Park, J. Y. Kim, J. H. Park, Physical review letters, **114**(11), 117603 (2015).
- [6] M. Staruch, J. F. Li, Y. Wang, D. Viehland, P. Finkel, Applied Physics Letters, **105**(15), 152902 (2014).
- [7] K. Rubi, P. Kumar, D. M. Repaka, R. Chen, J. S. Wang, R. Mahendiran, Applied Physics Letters, **104**(3), 032407 (2014).
- [8] N. N. Phuoc, C. K. Ong, Applied Physics Letters, **105**(2), 022905 (2014).
- [9] X. Wang, Z. Su, A. Sokolov, B. Hu, P. Andalib, Y. Chen, V. G. Harris, Applied Physics Letters, **105**(11), 112408 (2014).
- [10] Y. F. Duan, R. Zhang, C. Z. Dong, Y. Z. Luo, S. W. Or, Y. Zhao, K. Q. Fan, International Journal of Structural Stability and Dynamics, 1640016 (2015).
- [11] S. K. Mandal, G. Sreenivasulu, V. M. Petrov, G. Srinivasan, Applied Physics Letters, **96**(19), 192502 (2010).
- [12] S. C. Yang, C. W. Ahn, K. H. Cho, S. Priya, Journal of the American Ceramic Society, **94**(11), 3889 (2011).
- [13] S. C. Yang, K. H. Cho, C. S. Park, S. Priya, Applied Physics Letters, **99**(20), 202904 (2011).
- [14] Y. Zhou, S. C. Yang, D. J. Apo, D. Maurya, S. Priya, Applied Physics Letters, **101**(23), 232905 (2012).
- [15] Y. Zhou, S. Zhou, Y. Ye, L. Zhang, Optoelectron. Adv. Mater.-Rapid Comm., **6**(3-4), 401 (2012).
- [16] Z. Yun, C. Miao-Gen, F. Zhen-Jie, W. Xin-Yan, C. Yu-Jian, Z. Jin-Cang, Chinese Physics Letters, **28**(10), 107503 (2011).
- [17] Y. Zhou, J. Zhang, L. Li, Y. Su, J. Cheng, S. Cao, Journal of Alloys and Compounds, **484**(1), 535 (2009).

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