



Research articles

Stress-coefficient of magnetoelastic anisotropy in flexible Fe, Co and Ni thin films

Xiaoyuan Chen^{a,b,c}, Baomin Wang^{b,c,*}, Xingcheng Wen^{b,c}, Ping Sheng^{b,c},
Dhanapal Pravarthana^{b,c}, Huali Yang^{b,c}, Yali Xie^{b,c}, Haigang Liu^{d,e}, Xiaohong Xu^{a,*},
Run-Wei Li^{b,c,f,*}

^a Key Laboratory of Magnetic Molecules and Magnetic Information Materials of Ministry of Education, School of Chemistry and Materials Science, Shanxi Normal University, Linfen 041004, PR China

^b CAS Key Laboratory of Magnetic Materials and Devices, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201, PR China

^c Zhejiang Province Key Laboratory of Magnetic Materials and Application Technology, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201, PR China

^d Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201204, PR China

^e Shanghai Synchrotron Radiation Facility (SSRF), Shanghai 201204, PR China

^f Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, PR China

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ABSTRACT

Magnetoelastic anisotropy is one of the most essential and fundamental characteristics in the magnetic anisotropy, which holds the core application and broad prospects in advanced flexible magnetic devices. Here, we determined the stress-coefficient of magnetoelastic anisotropy of Fe, Co and Ni thin films deposited on flexible polyvinylidene fluoride (PVDF) substrate. Owing to an anisotropic thermal expansion of PVDF, a uniaxial stress can be effectively generated and transferred to the magnetic films in-situ by changing the temperature. The magnetic anisotropy constants at different compressive stresses were quantitatively investigated with anisotropic magnetoresistance (AMR). Through fitting the AMR curves at different compressive stresses, the stress-coefficient of magnetoelastic anisotropy of Fe, Co and Ni thin films were determined to be $(6.31 \pm 0.19) \times 10^3 \text{ erg cm}^{-3} \text{ GPa}^{-1}$, $(2.71 \pm 0.13) \times 10^4 \text{ erg cm}^{-3} \text{ GPa}^{-1}$ and $(2.46 \pm 0.19) \times 10^5 \text{ erg cm}^{-3} \text{ GPa}^{-1}$, respectively. These values are basic magnetic parameter for magnetic elements, which are helpful for evaluating the performance of magnetic devices under flexible/stretchable conditions.

1. Introduction

Magnetic anisotropy is an important physical parameter to determine the preferential alignment of spin orientations in magnetic materials [1,2], which is quite essential for various applications in many devices such as magnetic storage, magnetic sensors and non-volatile magnetic random access memories [3–5]. In particular, magnetoelastic anisotropy originates from the stress-induced magnetic anisotropy, which has received extensive attention recently for the high-speed development of flexible/stretchable magneto-electronic devices [6–13]. From the viewpoint of practical applications, it is necessary to understand the evolution of magnetoelastic anisotropy of magnetic thin films under stress, and to improve its performance of magneto-electronic

devices under flexible environment [14]. Therefore, to determine the ratio of change in magnetic anisotropy with stress, defined as stress-coefficient of magnetoelastic anisotropy, is of crucial significance not only to the fundamental magnetism but also for designing flexible magneto-electronic devices.

In this work, we determined the stress-coefficient of magnetoelastic anisotropy of Fe, Co and Ni thin films deposited on polyvinylidene fluoride (PVDF) substrate through anisotropic magnetoresistance (AMR) measurement. The PVDF substrate has an anisotropic thermal expansion, which can transfer a uniaxial compressive stress to the films grown on it merely by changing the temperature. Through fitting the AMR curves at different stresses, we calculated the stress-coefficient of magnetoelastic anisotropy of Fe, Co, and Ni thin films to be

* Corresponding authors at: CAS Key Laboratory of Magnetic Materials and Devices, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201, PR China.

E-mail addresses: wangbaomin@nimte.ac.cn (B. Wang), xuxh@sxnu.edu.cn (X. Xu), runweili@nimte.ac.cn (R.-W. Li).

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$(6.31 \pm 0.19) \times 10^3 \text{ erg cm}^{-3} \text{ GPa}^{-1}$, $(2.71 \pm 0.13) \times 10^4 \text{ erg cm}^{-3} \text{ GPa}^{-1}$ and $(2.46 \pm 0.19) \times 10^5 \text{ erg cm}^{-3} \text{ GPa}^{-1}$, respectively.

2. Experimental method

Ta(2 nm)/Fe (Co or Ni) (15 nm)/Ta(2 nm) were deposited on 50 μm thick PVDF and Si substrates by magnetron sputtering at room temperature. The thickness of 2 nm Ta film deposition on the bottom (top) of the magnetic thin films were employed to reduce the roughness of flexible substrate and prevent the sample from oxidization, respectively. Thermomechanical analyzer was utilized to determine the anisotropic thermal expansion coefficient of PVDF, which is deduced to be $\alpha_{31} = 150 \times 10^{-6} \text{ K}^{-1}$ and $\alpha_{32} = 23.2 \times 10^{-6} \text{ K}^{-1}$ in the temperature range of 200 to 300 K [15]. The magnetic hysteresis (MH) loops and AMR curves under different stresses are measured by superconducting quantum interference device (SQUID) and physical property measurement system (PPMS).

3. Results and discussion

3.1. Thermal expansion of PVDF and magnetoelastic anisotropy of magnetic materials

Fig. 1(a) shows a schematic of an anisotropic thermal expansion of PVDF substrate, where α_{31} and α_{32} is defined as the thermal expansion coefficient along x and y directions, respectively. The α_{31} is greater than α_{32} by more than an order of magnitude, which means the shrinkage of PVDF along the x direction is greater than y direction. So, a uniaxial compressive stress can be generated on lowering the temperature, and transferred to the magnetic films grown on the top of PVDF that results in magnetoelastic anisotropy. In order to give a more intuitive understanding of the magnetoelastic anisotropy, the different scenarios of stress induced magnetic anisotropy is described in Fig. 1(b). The zero magnetoelastic anisotropy is defined as the magnetic easy axis, which is not sensitive to stress. The positive (negative) magnetoelastic anisotropy is defined as the magnetic easy axis prefers to be perpendicular (parallel) to the applied compression stress.

3.2. Stress dependence of magnetic hysteresis loop

To investigate the stress induced magnetic anisotropy of Fe, Co and Ni films on PVDF substrate. The MH loops of the films were measured as a function of temperature along x and y directions. The stress induced in the films at different temperature due to an anisotropic thermal expansion of PVDF substrate can be given as [7,15–17]

$$\sigma = \varepsilon E_f / (1 - \nu^2) \quad (2)$$

where E_f is the Young's modulus of Fe ($\sim 131 \text{ GPa}$), Co ($\sim 114 \text{ GPa}$) and Ni ($\sim 133 \text{ GPa}$), ν is the Poisson ratio of Fe (~ 0.37), Co (~ 0.40) and Ni (~ 0.38) films [18], ε is the effective strain along x direction, which can be written as [15]

$$\varepsilon = \Delta T (\alpha_{32} - \alpha_{31}) \quad (3)$$

Fig. 2(a)–(f) compares the MH loops of Fe, Co and Ni films on PVDF along x and y directions under different compressive stresses. The normalized MH loops display notable and robust changes in the presence of compressive stress. The MH loops of Co film become more slanted (squarer) along x (y) direction with the increase of compressive stress [Fig. 2(b) and (e)]. However, the MH loops of Fe and Ni films along x and y directions show an opposite behavior with the increase of compressive stress [Fig. 2(a), (d) and (e), (f)]. The compressive stress dependence of normalized remnant magnetization (M_r/M_s) for Fe, Co and Ni films is summarized in Fig. 2(g)–(i). For Co films, the M_r/M_s decreases (increases) along x (y) directions as shown in Fig. 2(h). While for Fe and Ni films, the M_r/M_s increases (decreases) along x (y) directions as shown in Fig. 2(g) and (i), respectively. The obtained results are different from that observed in the films on the Si substrate with different temperatures as shown in Fig. 3, which indicates that enhanced magnetic anisotropy is induced by anisotropic thermal expansion of the PVDF substrate. The reason why Fe and Ni films reveal the opposite behavior in comparison with Co films under similar compressive stress conditions is that Co owns the positive magnetostriction coefficient while the negative magnetostriction coefficient for Fe and Ni [2]. Further, it can be observed that the M_r/M_s increases stronger along the easy axis in the order of Fe, Co, and Ni thin films as shown in Fig. 2(g), (h), and (i), respectively. This phenomenon can also be proved by the stress-coefficient of magnetoelastic anisotropy, which will be discussed in the last section.

3.3. Determination of magnetic anisotropy constant

To investigate the magnetoelastic anisotropy quantitatively, the magnetic anisotropy constants of Fe, Co and Ni films under different compressive stresses were determined by using the AMR measurement. The AMR curves were measured by using the standard four-probe technique by changing the angle θ between the magnetic field and current (I) from 0 to 360° in the PVDF-based magnetic films while I is fixed along either x or y direction, respectively [Inset of Fig. 4(a) and (b)]. The representative curves for normalized magnetic torque which is the relationship of the angle between the magnetization and magnetic field can be obtained. The AMR equation is given as [19–24]

$$R_{xx} = R_{\perp} + (R_{\parallel} - R_{\perp}) \cos^2 \theta_M \quad (1)$$

where θ_M is the angle between magnetic moment (M) and I directions,

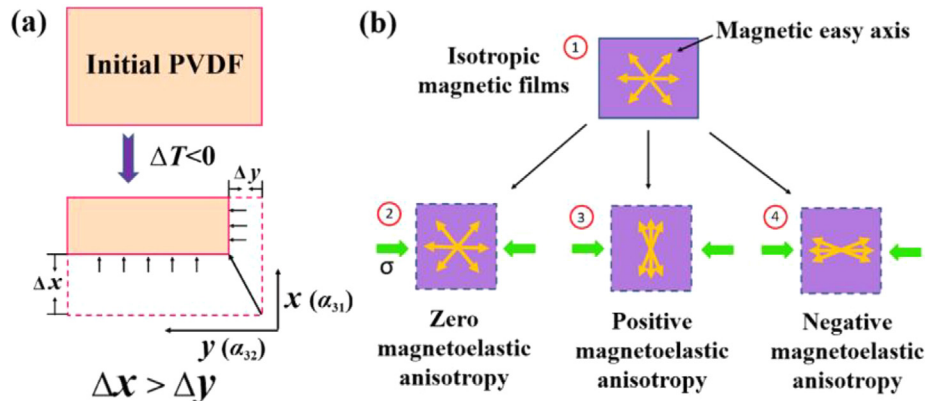


Fig. 1. (a) The schematic diagram of PVDF deformation along α_{31} and α_{32} directions during the lowering of temperature from room temperature. (b) Classification of magnetoelastic anisotropy (under compressive stress): zero magnetoelastic anisotropy positive magnetoelastic anisotropy and negative magnetoelastic anisotropy.

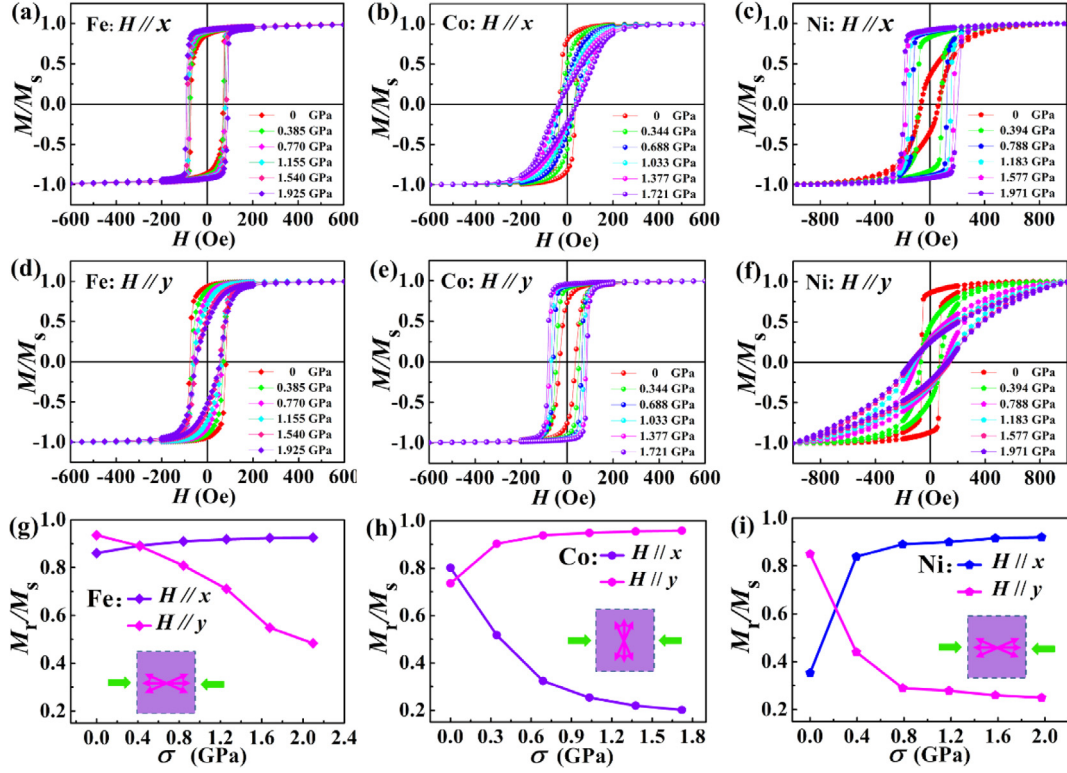


Fig. 2. (a)-(f) The normalized MH loops under different compressive stress in Fe, Co and Ni films along x and y directions on PVDF substrate, respectively. (g)-(i) The compressive stress dependence of M_r/M_s along x and y direction in Fe, Co and Ni, respectively. The inset shows the applied compressive stress configuration with respect to the initial easy axis.

and the resistance of minimum value $R_{//}$ and the maximum R_{\perp} , where $//$ and \perp corresponds to H parallel and perpendicular to I direction, respectively.

Considering the Co films on PVDF for example, the curves of AMR follow the orientation of external field that shows a periodically smooth behavior at 240 K for I along x and y directions in Fig. 4(a) and (b), respectively. The solid curves are $\cos^2\theta$ dependent, which coincides with the AMR curve under 8000 Oe, indicates that the field is large enough to ensure the magnetic moments align along the field for coherent rotation without hysteresis, which also implies the $\theta_M = \theta_H$, where θ_H is the angle between the direction of H and I . But magnetic moment M_{Co} can no longer follow the $\cos^2\theta_H$ relationship due to presence of magnetic anisotropy when the applied magnetic field is less than the saturated field. This results in the $\theta_M \neq \theta_H$ from 0 to 360° when I along x and y directions.

However, the AMR values are directly related to θ_M on the basis of Eq. (1). According to the different relationship of θ_M and θ_H , we can further calculate and compare magnetic torque at different fields by using the normalized magnetic torque equation, which can be expressed as

$$l(\theta_M) = L(\theta_M)/\mu_0 M_s H = \sin(\theta_H - \theta_M) \quad (2)$$

As shown in Fig. 5(a) and (b), normalized magnetic torque curves exhibits different profile and increases with the decrease in the applied magnetic field for I along x and y directions, respectively. When the magnetic field is applied greater or equal to a saturation field, the magnetic torque curves shows a constant profile that indicates the effect of magnetic torque is negligible. While the effect of magnetic torque is much more pronounced when the applied magnetic field is lower than saturation field. Besides, we can witness that the experimental and simulated normalized magnetic torque curves can overlap well, implying that magnetic torque can be calculated accurately by Eq. (2).

During the test of magnetic torque, the magnetization-reversal process is largely governed by the symmetry and anisotropic energies, which is related to anisotropic constant. So, the free energy density of the magnetic films with external field can be written as [25]

$$E = K_u \sin^2 \theta_M - \mu_0 M_s H \cos(\theta_H - \theta_M) \quad (3)$$

In the equilibrium state $\frac{\partial E}{\partial \theta_M} = 0$, the normalized magnetic torque can be written as

$$l(\theta_M) = \sin(\theta_H - \theta_M) = [K_u/(\mu_0 M H)] \sin(2\theta_M) \quad (4)$$

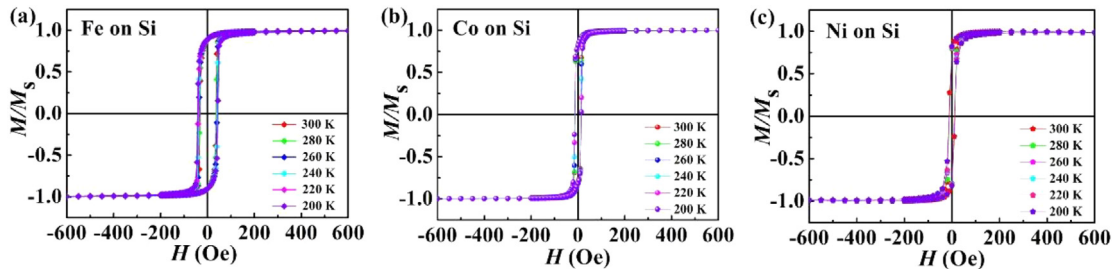


Fig. 3. The normalized MH loops under different temperatures in Fe (a), Co (b) and Ni (c) films on Si substrate.

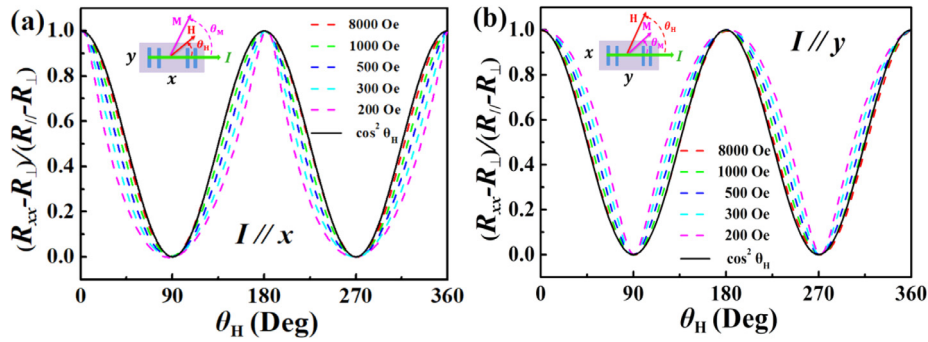


Fig. 4. The angular dependence of AMR curves at different magnetic fields in Co films for I is along x (a) and y (b) directions at 240 K.

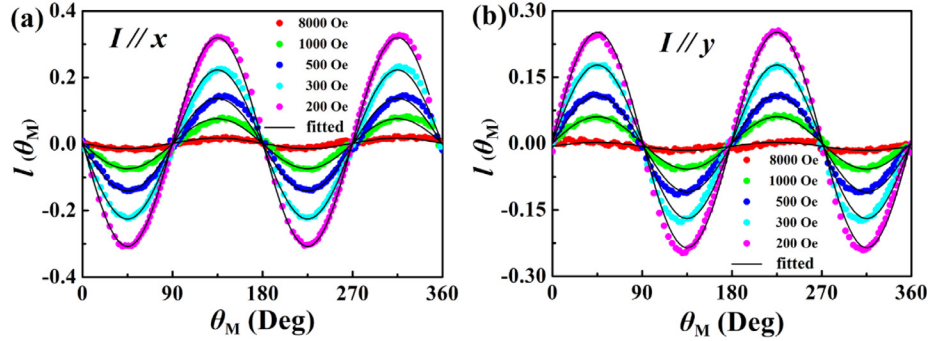


Fig. 5. The experimental and simulated normalized magnetic torque curves in Co films under different magnetic fields for I along x (a) and y (b) directions at 240 K.

where E is the anisotropy energy, μ_0 is the magnetic permeability, M_s is the saturation magnetization, and K_u is the magnetic anisotropy constant. Thereby, we can obtain the K_u values under different magnetic fields by fitting the magnetic torque curves by Eq. (4) in Fig. 5(a) and (b).

3.4. Determination of stress-coefficient of magnetoelastic anisotropy

Fig. 6(a)–(f) shows the compressive stress dependence of the normalized magnetic torque curves of Fe, Co and Ni films for I along x and y directions, respectively. It clearly shows that the uniaxial magnetic anisotropy is enhanced with the increase of applied compressive stress. Fe and Ni films display an opposite phenomenon for I along x and y

directions in comparison with Co films, which is consistent with the negative magnetoelastic anisotropy of Fe and Ni observed in Fig. 2. By employing the normalized magnetic torque curves in Fig. 6, the K_u values of Fe, Co and Ni films can be calculated by Eq. (4). As it can be seen from the fitted outcomes in Fig. 7, K_u increases with the increase of applied compressive stress. The change in the magnetic anisotropy of the Fe, Co and Ni thin films on PVDF can be mainly attributed to the change of magnetoelastic anisotropy. This also evidence by no change in magnetic anisotropy of the films on Si within the measured temperature range, because Si substrate does not provide anisotropy thermal expansion like observed in PVDF substrate. The computed stress-coefficient of magnetoelastic anisotropy ($\Delta K_u/\Delta\sigma$) for Fe, Co, and Ni films are $(6.31 \pm 0.19) \times 10^3 \text{ erg cm}^{-3} \text{ GPa}^{-1}$,

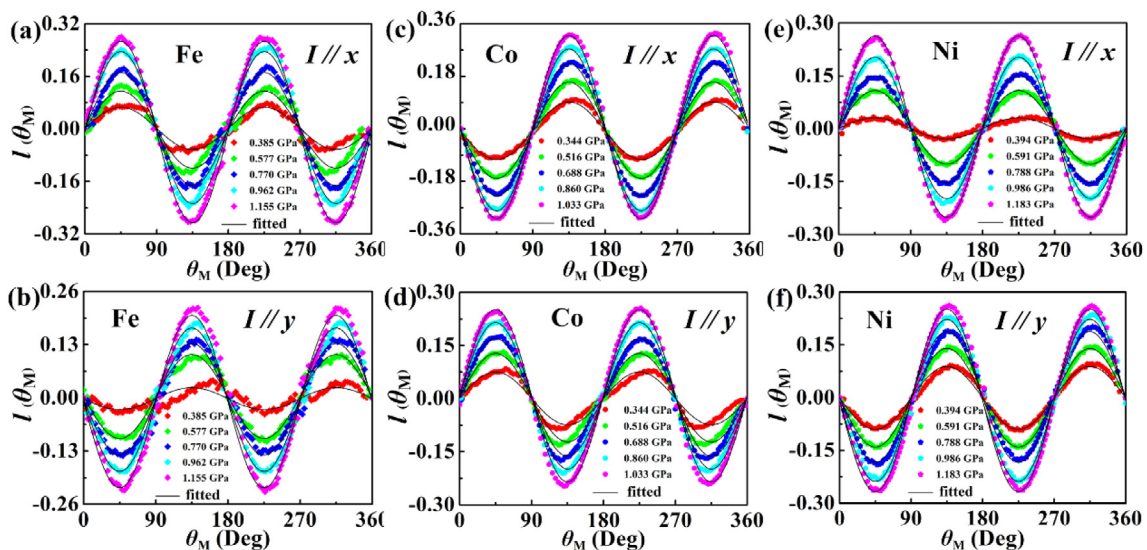


Fig. 6. The experimental and fitted normalized magnetic torque curves under different compressive stresses for I along x and y directions, respectively, in Fe (a,b), Co (c,d), and Ni (e,f) films.

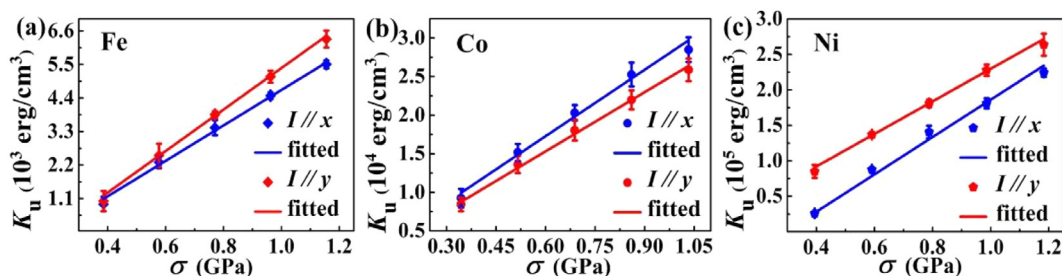


Fig. 7. The K_u as the function of the compressive stress for I along x and y directions, respectively, in Fe (a), Co (b) and Ni (c) films.

$(2.71 \pm 0.13) \times 10^4 \text{ erg cm}^{-3} \text{ GPa}^{-1}$ and $(2.46 \pm 0.19) \times 10^5 \text{ erg cm}^{-3} \text{ GPa}^{-1}$, respectively. These are the average values from AMR measurement for I along x and y directions, respectively. The stress-coefficient of magnetoelastic anisotropy shows a significant increase in the trend by an order of magnitude with the ranking of Fe, Co and Ni, which is consistent with discussed in the previous section.

4. Conclusions

In summary, we have systematically investigated the magnetoelastic anisotropy of magnetic elements Fe, Co, and Ni thin films deposited on PVDF substrate. From the MH loops analysis under various compressive stresses, it is revealed qualitatively that Co thin film exhibits positive magnetoelastic anisotropy, while Fe and Ni thin films have negative magnetoelastic anisotropy. The magnetoelastic anisotropy constants for these thin films under different compressive stresses were determined using AMR measurements. Thus, the stress-coefficient of magnetoelastic anisotropy of Fe, Co, and Ni thin films was determined to be $(6.31 \pm 0.19) \times 10^3 \text{ erg cm}^{-3} \text{ GPa}^{-1}$, $(2.71 \pm 0.13) \times 10^4 \text{ erg cm}^{-3} \text{ GPa}^{-1}$ and $(2.46 \pm 0.19) \times 10^5 \text{ erg cm}^{-3} \text{ GPa}^{-1}$, respectively. The determination of this basic magnetic parameter for magnetic elements is helpful for evaluating the performance of magnetic devices under flexible/stretchable conditions.

CRediT authorship contribution statement

Xiaoyuan Chen: Investigation, Formal analysis, Writing - original draft. **Baomin Wang:** Conceptualization, Writing - review & editing, Supervision, Funding acquisition. **Xingcheng Wen:** Methodology. **Ping Sheng:** Investigation. **Dhanapal Pravarthana:** Writing - review & editing. **Huali Yang:** Formal analysis. **Yali Xie:** Formal analysis. **Haigang Liu:** Methodology. **Xiaohong Xu:** Supervision, Funding acquisition. **Run-Wei Li:** Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://>

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