TOPICAL REVIEW — Magnetism, magnetic materials, and interdisciplinary research

Fabrication, properties, and applications of flexible magnetic films*

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Flexible magnetic devices, i.e., magnetic devices fabricated on flexible substrates, are very attractive in applications such as detection of magnetic field in an arbitrary surface, non-contact actuators, and microwave devices, due to their stretchable, biocompatible, light-weight, portable, and low cost properties. Flexible magnetic films are essential for the realization of various functionalities of flexible magnetic devices. To give a comprehensive understanding for flexible magnetic films and related devices, recent advances in the study of flexible magnetic films are reviewed, including fabrication methods, magnetic and transport properties of flexible magnetic films, and their applications in magnetic sensors, actuators, and microwave devices. Our aim is to foster a comprehensive understanding of these films and devices. Three typical methods have been introduced to prepare the flexible magnetic films, by deposition of magnetic films on flexible substrates, by a transfer and bonding approach or by including and then removing sacrificial layers. Stretching or bending the magnetic films is a good way to apply mechanical strain to them, so that magnetic anisotropy, exchange bias, coercivity, and magnetoresistance can be effectively manipulated. Finally, a series of examples is shown to demonstrate the great potential of flexible magnetic films for future applications.

Keywords: flexible, magnetic films, strain

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1. Introduction

Silicon wafers have been widely used in manufacturing electronic devices.[1] However, the wafers are rigid, while many natural things related to applications, such as human bodies, organisms, clothes, etc., are elastic, soft, and curved.[2] Therefore, electronic devices based on rigid silicon wafers are not suitable for these up and coming applications. Flexible electronics is a technology for assembling electronic circuits and devices on flexible substrates, which are significantly of lower cost, lighter, and more compact, as compared to the conventional electronic devices.[3] By thinning a single crystal silicon wafer to 100 μm, the first flexible solar cell was made in 1960,[4,5] which started the development of flexible electronics. In 1997, polycrystalline silicon thin film transistors (TFT) made on plastic substrates were reported and received lots of attention because of potential applications in flexible displays.[6,7] Since then, the flexible electronics field has developed rapidly, and commercialized products have appeared in our daily life. Now, flexible electronic devices are widely used in displays, radio-frequency identification (RFID), solar cells, lighting, and sensors, among which flexible displays occupy more than 80% of the market for flexible electronics.[8–10] With the development of various flexible devices, displays, logic, sensors, and memory units are expected to be integrated in a multi-functional system fabricated on a flexible substrate.[11]

It is well known that magnetic materials are important in the fabrication of electronic devices.[12] For example, soft magnetic materials are usually applied in inductors, transformers, microwave devices, and screening of magnetic fields.[13,14] Hard magnetic materials are widely used in loudspeakers, generators, memory units, and sensors.[15] Magnetostrictive materials are used in actuators.[16] Permalloy can be used in anisotropic magnetoresistance (AMR) sensors.[17] Giant magnetoresistance (GMR) or tunneling magnetoresistance (TMR) multilayered structures can be used in high-speed read heads in disk memory devices owing to their large magnetoresistance.[18,19] Recently, GMR or TMR sensors fabricated on flexible substrates, so-called flexible magnetoelectronics, have attracted a lot of interest due to their potential applications in detecting magnetic fields in living organisms.[20,21] Because of the extensive applications of magnetic materials, integration of flexible magnetic materials in flexible electronics is inevitable.

In this review, we first introduce the techniques of fabricating flexible magnetic films and devices. Then we focus on the properties of flexible magnetic films and devices. Finally, the applications of flexible magnetic films and devices are discussed.

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2. Fabrication of flexible magnetic films

2.1. Magnetic films deposited on flexible substrates

Fabricating magnetic films directly on flexible substrates is a straightforward way to get flexible magnetic films. The most used flexible substrates are organic polymers including polyethylene terephthalate (PET), polyethylene naphthalate (PEN), polyethersulphone (PES), polyimide (PI), and polydimethylsiloxane (PDMS).[22] These organic polymers are highly flexible, inexpensive, and compatible with roll-to-roll processing.[23] Most polymers cannot tolerate high-temperature treatment,[24] but they are still suitable for fabrication of most magnetic films and devices because of the near-room temperature deposition and low-temperature (usually lower than 400 °C) post-annealing.

For fabricating magnetic films on flexible substrates, a suitable buffer layer is often required to reduce the roughness of flexible substrates and ensure the continuity and functionality of magnetic films and multilayered structures, such as giant magnetoresistance and spin-valve devices.[25–27] For example, the root-mean-square (RMS) roughness of a PET substrate is about 2.16 nm, which is much larger than that of the thermally oxidized Si substrate. The 150-nm-thick $Fe_{81}Ga_{19}$ film grown directly on flexible PET substrates exhibits a root-mean-square roughness of 3.34 nm.[28] A 30-nm-thick Ta buffer layer can reduce the roughness of $Fe_{81}Ga_{19}$/Ta/PET films to 2.04 nm.[29] The flexible exchange-biased Ta(5 nm)/$Fe_{81}Ga_{19}$(10 nm)/$Ir_{20}Mn_{80}$(20 nm)/Ta(30 nm)/PET heterostructures have been successfully fabricated by growing a Ta buffer layer, which could be used to stabilize the magnetization of magnetic layers in flexible spin-valve devices.[30] Chen *et al.* have fabricated flexible Co/Cu GMR multilayers on polyester substrates by DC magnetron sputtering.[31] The sample structure is schematically shown in Fig. 1(a). Figure 1(b) shows the photographic image of circularly bent Co/Cu multilayer deposited on polyester substrate. Before deposition of the Co/Cu multilayer, AR-P 3510 positive photoresist (Allresist, Germany) buffer layer with a thickness of 2 μm is spin-coated on flexible substrates to reduce the surface roughness. Besides, Oh *et al.* fabricated flexible spin-valve structures of Ta (3 nm)/NiFe (10 nm)/Cu (1.2 nm)/NiFe (3 nm)/IrMn (10 nm)/Ta (3 nm) on PEN substrates using AZ 5214E photoresist as buffer layers.[32] Melzer *et al.* provided another way to fabricate flexible spin-valve structures, as shown in Fig. 2.[33] First, PDMS was spin-coated onto silicon wafers with surface roughness < 0.5 nm. Then, the structure of Ta (2 nm)/IrMn (5 nm)/ [Permalloy (Py) (4 nm)/CoFe (1 nm)]/Cu (1.8 nm)/[CoFe (1 nm)/Py (4 nm)] with 5-nm-thick Ta buffer layer was fabricated by magnetron sputtering. After a lithographic lift-off process, by means of an antistick layer, the PDMS film is peeled from the rigid silicon wafer, forming a flexible magnetic multilayered structure.

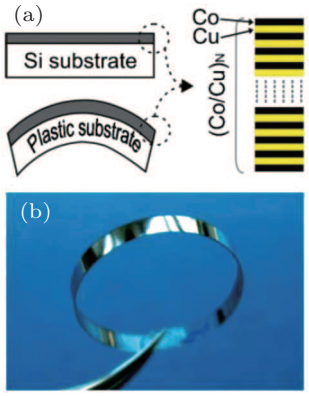


Fig. 1. (a) Schematic illustration of $(Co/Cu)_N$ films deposited on Si and flexible substrates. (b) Photographic image of circularly bent $(Co/Cu)_{20}$ film deposited on polyester substrate.[31]

With the development of magnetoelectric materials, multiferroic composites consisting of magnetostrictive materials and organic ferroelectric materials — such as polyvinylidene fluoride (PVDF) and polyvinylidene-fluoride–trifluoroethylene (PVDF-TrFE) — have received much attention.[34] Heterostructural Sm–Fe/PVDF films have been prepared by depositing Sm–Fe nanoclusters onto flexible PVDF membranes using cluster beam deposition, which exhibits large magnetoelectric voltage output of 210 μV at an external magnetic bias of 2.3 kOe (1 Oe = 79.5775 $A \cdot m^{-1}$).[34]

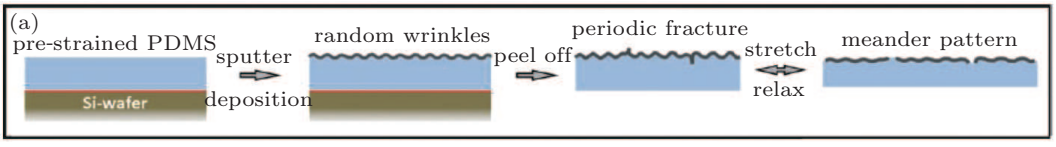


Fig. 2. Fabrication process of stretchable spin-valve structure.[33]

Besides organic polymers, people also seek other types of flexible substrates to prepare magnetic films. Liang *et al.* have successfully produced flexible graphene/Fe_3O_4 hybrid papers by using graphene as flexible substrates, as shown in Fig. 3.[35] During fabrication, a two-step process was used: (I) mixing graphene aqueous solution with water-soluble Fe_3O_4 nanoparticles and (II) chemical reduction of the suspension of water-soluble Fe_3O_4 nanoparticles and graphene sheets with

hydrazine. In a broad sense, cantilevers several micrometers thick, which have been applied as sensors in micro-electro-mechanical systems (MEMS),[36] can be employed as flexible substrates. The cantilevers are usually made of materials including Si, polyimide, Si_3N_4, etc.[37] Onuta *et al.* have fabricated a flexible multiferroic composite as an energy harvester consisting of a magnetostrictive $Fe_{0.7}Ga_{0.3}$ thin film and a $Pb(Zr_{0.52}Ti_{0.48})O_3$ piezoelectric thin film on a 3.8-μm-thick Si cantilever, as shown in Fig. 4.[38]

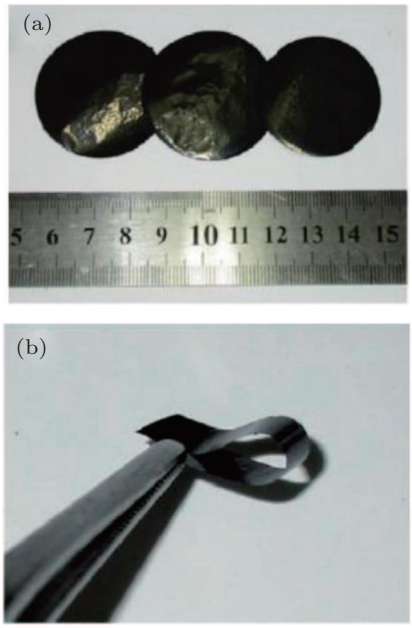


Fig. 3. Flexible graphene/Fe_3O_4 hybrid papers using graphene as flexible substrates.[35]

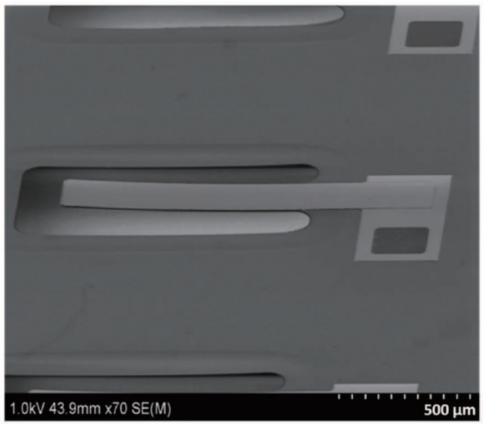


Fig. 4. Scanning electron micrograph (SEM) of a flexible multiferroic energy harvester.[38]

Magnetic polymers fabricated by dispersing magnetic components in a polymer matrix can serve as flexible functional cantilevers that respond to external magnetic fields and mechanical vibrations. If the magnetic components are magnetostrictive materials and the polymers are ferroelectric materials, three main types of polymer-based multiferroic materials can be achieved: nano-composites, laminated composites, and polymer as a binder composite, shown in Figs. 5(a), 5(b), and 5(c), respectively.[39] The investigations of polymer-based multiferroic materials are challenging and innovative, bridging the gap between fundamental research and near-future applications.

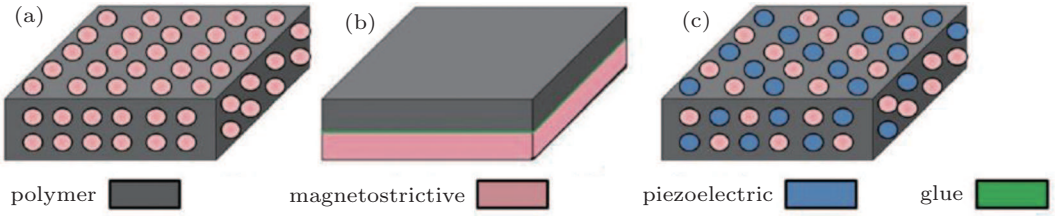


Fig. 5. The three main types of polymer-based multiferroic materials.[39]

2.2. Transfer and bonding approach

In the transfer and bonding approach, first, magnetic films or structures are fabricated on conventional rigid substrates like Si wafer, glass, MgO, etc., by standard fabrication methods. Then the magnetic films or structures can be transferred by removing the substrates through laser annealing, chemical solution, or directly peeling off the films.[40–42] Finally, the transferred films or structures can be bonded to flexible substrates by glue or physical/chemical adsorption.[40–42] Generally, conventional rigid substrates are much flatter than flexible substrates. Therefore, the transfer and bonding approach can provide flexible magnetic films and structures with high quality and high performance. However, it is difficult to prepare a large-area flexible film by this method due to random damage during the transfer procedure. Since the discovery of graphene, a variety of transfer and bonding approaches have been developed to transfer graphene films onto flexible substrates.[43] However, these methods cannot be directly employed to prepare flexible magnetic films, because some chemicals used in the transfer procedure may damage the magnetic films.[44] Donolato *et al.* have developed another innovative, simple, and versatile pathway to trans-

fer magnetic films and structures onto flexible polymer substrates, as shown in Fig. 6.[45] At first, a trilayered structure of Ti/Au/SiO_2, which acts as the donor substrate, was grown on a conventional Si substrate using a sputter deposition system.

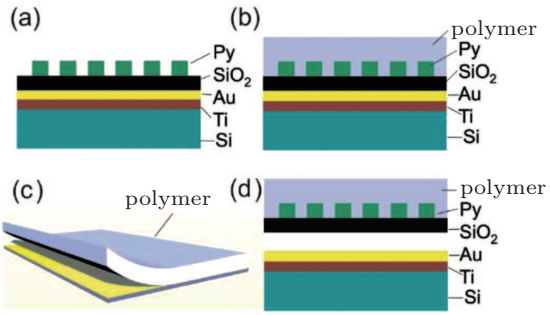


Fig. 6. Schematic diagram of the transfer and bonding approach to get flexible magnetic films.[45]

Then, magnetic nanostructures made of permalloy ($Ni_{80}Fe_{20}$) were fabricated on the trilayer by means of an electron beam lithography process. After that, PDMS was spin-coated on the magnetic nanostructure. Finally, the transfer was accomplished by simple immersion of the chip in water and a gentle mechanical lifting of the polymer membrane off the substrate. In this approach, the selection of Au and SiO_2 layers is key. Due to the hydrophobic character of Au and hydrophilic behavior of SiO_2, the bond between them is rather weak, so the Au and SiO_2 layers can be easily separated via the water-assisted lift-off process.

2.3. Release of sacrificial layers

The general processes of releasing sacrificial layers for preparing flexible magnetic films are shown in Fig. 7. Magnetic films are first deposited on bulk substrates or bulk substrates with sacrificial layers. For the simple film/substrate structures, the substrates themselves can be treated as sacrificial layers. The substrates or the sacrificial layers can be removed by an aqueous solution of chemicals, chemical etching, or dry etching to achieve freestanding magnetic films.[46,47] This method has the advantages similar to the above-mentioned transfer and bonding approach and can provide flexible magnetic films with high quality.

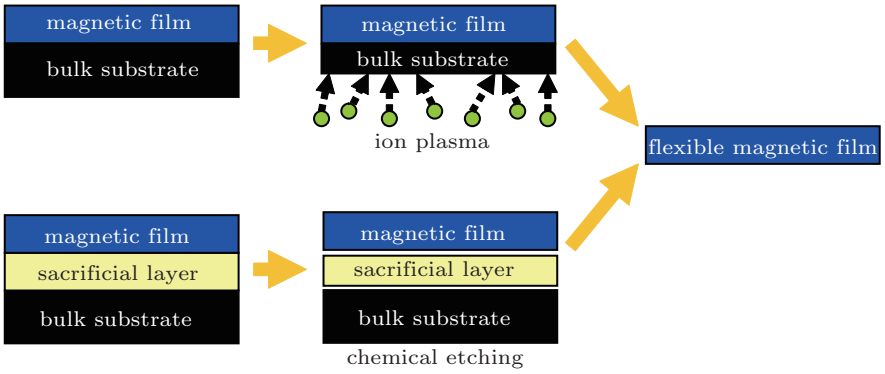


Fig. 7. General processes of releasing sacrificial layers in preparing flexible magnetic films.

The most used sacrificial layers are NaCl and photoresist. Heczko and Thomas have epitaxially grown Ni–Mn–Ga films on water-soluble (001)-oriented NaCl single crystals and obtained high quality free-standing Ni–Mn–Ga films by dissolving the NaCl substrates.[48] In contrast, Tillier *et al.* have used photoresist as sacrificial layers to fabricate flexible Ni–Mn–Ga films.[49] Ni–Mn–Ga films were first deposited on photoresist (Shipley S1818) layers that had been spin-coated on polycrystalline Al_2O_3 substrates. After deposition, the samples were placed in an acetone bath to remove the photoresist sacrificial layer and obtain freestanding magnetic films. Other materials, such as Au, MgO, and Cr, can also be used as sacrificial layers due to their chemical soluble properties.[50–52] For example, Bechtold *et al.* have prepared 1.2-μm-thick $Fe_{70}Pd_{30}$ films on Au(50 nm)/Cr(8 nm)/MgO substrates.[50] After deposition, the $Fe_{70}Pd_{30}$ films were released from the substrate by wet chemical etching of the sacrificial Au layer in an aqueous solution of potassium iodide and iodine. Although removing sacrificial layers is a good way to prepare high quality flexible magnetic films, this method still has some disadvantages. For example, the aqueous solution of chemicals may damage the magnetic films. Alternatively, flexible membranes of inert materials, such as Pt and Au, can be prepared by removing sacrificial layers. Then magnetic films can be deposited on the thin metallic membranes. We have etched platinized Si substrates (Pt(200 nm)/Ti(50 nm)/SiO_2(500 nm)/Si) in 10 wt% HF solutions for 4 h.[53] Because the Ti and SiO_2 layers reacted with HF, producing soluble SiF_4 and TiF_3, respectively, the 200-nm-thick Pt layers were released from the Si substrates. The resulting flexible Pt foils can be used as flexible substrates for preparing ferromagnetic or ferroelectric films at an elevated temperature. The detailed processes are shown schematically in Fig. 8.

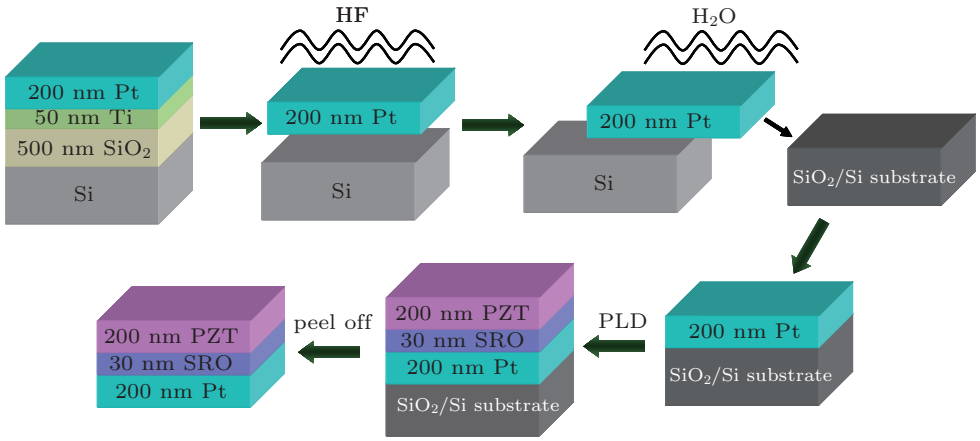


Fig. 8. Method to obtain flexible functional thin-films using Pt foils.[53]

3. Properties of flexible magnetic films

3.1. Effect of buffer layer

Prior to fabricating flexible magnetic films and devices, an appropriate buffer layer needs to be introduced to decrease the roughness of flexible substrates, improve the crystal orientation of magnetic films, and release residual stress. Therefore, the buffer layers are extremely important in determining the properties of flexible magnetic films, such as magnetic anisotropy, coercivity, magnetoresistance, etc.[54] We have investigated the effect of a Ta buffer layer on the magnetic properties of magnetostrictive $Fe_{81}Ga_{19}$ films grown on flexible PET substrates.[29] As shown in Fig. 9, with increasing the thickness of Ta buffer layer, both the uniaxial magnetic anisotropy and the coercivity of $Fe_{81}Ga_{19}$/Ta/PET films are decreased. Obviously, the Ta buffer layer releases the residual stress in PET substrates and therefore reduces the strength of the uniaxial anisotropy of $Fe_{81}Ga_{19}$ layers. The decrease of coercivity of $Fe_{81}Ga_{19}$ films may result from both the decrease of uniaxial anisotropy and the flatness of the films. Chen *et al.* have shown that the GMR effect of Co/Cu multilayers (MLs) on a flexible organic substrate can be enhanced up to 200% by introducing a photoresist (PR) buffer layer to flatten the plastic substrates.[31] They compared three Co/Cu multilayers grown on Si substrate, polyester substrate (P), and polyester substrate with a 2-μm photoresist (PR) buffer layer (PR+P). As shown in Fig. 10(d), the RMS roughness, R_q, of the P substrate is one order of magnitude larger than that of the Si substrate, and R_q of the PR+P substrate is close to that obtained for the Si substrate. As shown in Figs. 10(a), 10(b), and 10(c), GMR values significantly increase after introducing a PR buffer layer and rise to even higher values than those achieved on Si substrates due to an increased antiferromagnetic coupling fraction of the flexible PR buffered Co/Cu multilayers.

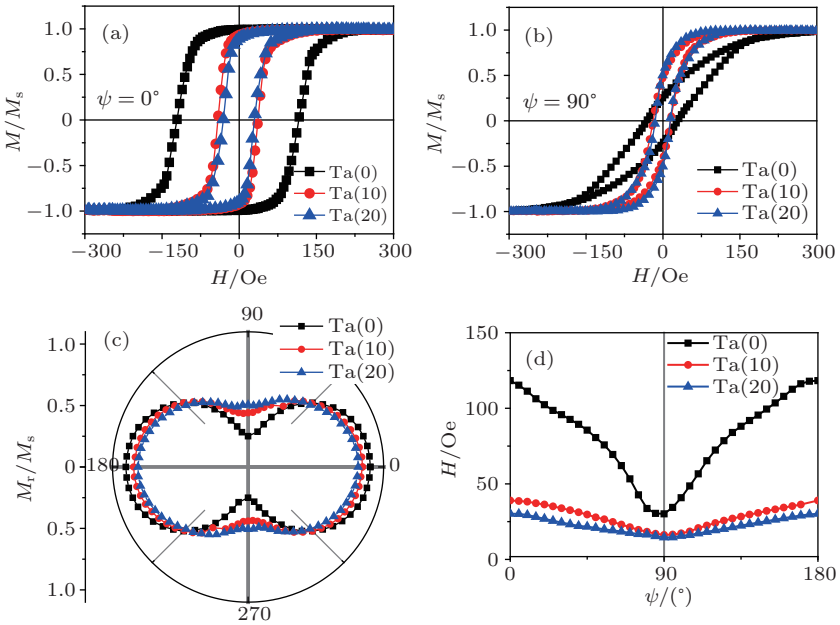


Fig. 9. Hysteresis loops for flexible $Fe_{81}Ga_{19}$(50 nm)/Ta/PET films with a magnetic field applied along (a) the easy ($\psi = 0°$) and (b) hard axes ($\psi = 90°$), and the corresponding angular dependence of (c) squareness and (d) coercive field. Ta(0), Ta(10), and Ta(20) indicate Ta buffer layer with thicknesses of 0, 10, and 20 nm, respectively.[29]

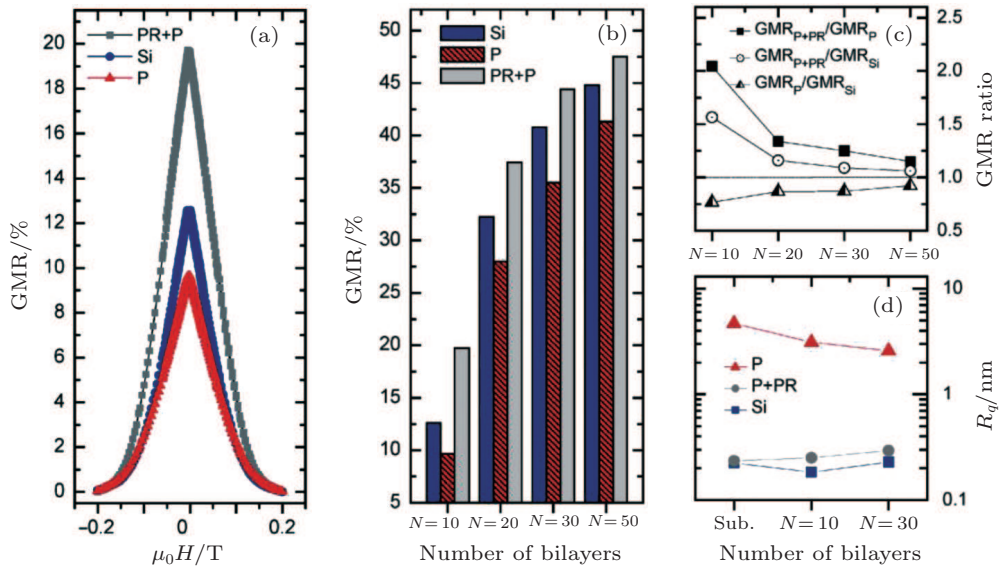


Fig. 10. (a) GMR curves of $(Co/Cu)_{10}$ MLs deposited on Si, P, and PR+R substrates. (b) GMR comparison of $(Co/Cu)_N$ MLs on various substrates with different numbers of bilayers. (c) GMR ratio of $(Co/Cu)_N$ MLs deposited on Si, P, and PR+P substrates. (d) RMS roughness, R_q, of Si, P, PR+P bare substrate and Co/Cu films on corresponding substrates.[31]

3.2. Strain dependence of magnetic properties

Control of the magnetic properties of flexible magnetic films via mechanical strains is an interesting topic from the viewpoint of both fundamental researches and potential applications.[55] Magnetic anisotropy is a key characteristic in determining the direction of magnetization and affecting the performance of spintronic devices.[56] For flexible spintronic devices applied in curved surfaces or used to evaluate the mechanical strain, their magnetic anisotropy under various mechanical strains need to be known and well controlled. $Fe_{1-x}Ga_x$ magnetostrictive alloy exhibiting moderate magnetostriction ($\sim$ 350 ppm for Ga content of 19%) under very a low magnetic field ($\sim$ 100 Oe) but good mechanical properties is a potential material applied in strain controllable spintronic devices. We have fabricated magnetostrictive $Fe_{81}Ga_{19}$ films on flexible PET substrates.[28] Due to the residual stress of the flexible substrates, a uniaxial magnetic anisotropy is observed for the as-grown $Fe_{81}Ga_{19}$ films. By inward or outward bending of the PET substrates, compressive and tensile strains can be applied to the $Fe_{81}Ga_{19}$ films. The hysteresis loops for $Fe_{81}Ga_{19}$/PET films under tensile and compressive strains are measured by bending the substrates along the easy or hard axis of the $Fe_{81}Ga_{19}$ films, as shown in Fig. 11.[28] For the magnetic field oriented along the easy axis, a tensile strain along the hard axis gives rise to a drastic decrease in M_r/M_s ratio, as shown in Fig. 11(a). In contrast, under a compressive strain, the M_r/M_s ratio is increased, as shown in Fig. 11(b). For the magnetic field oriented along the hard axis, the M_r/M_s ratio is decreased and increased under a tensile and compressive strain applied along the easy axis, respectively, as shown in Figs. 11(c) and 11(d). The results provide an alternative way to mechanically tune magnetic properties, which is particularly important for developing flexible magnetic devices.

In addition, we have also studied the effect of mechanical strain on magnetic properties of flexible exchange biased $Fe_{81}Ga_{19}$/IrMn heterostructures grown on PET substrates.[57] Figure 12 shows typical results for the in-plane strain dependence of the normalized magnetic hysteresis loops of $Fe_{81}Ga_{19}$(10 nm)/IrMn(20 nm) bilayers with strain applied perpendicular or parallel to the pinning direction (PD). The exchange bias field achieves a maximum value of 69 Oe for magnetic field applied along the induced PD and vanishes for magnetic field perpendicular to the PD, as shown in Figs. 12(a) and 12(b). The loop squareness decreases when a tensile strain is applied perpendicular to the PD with magnetic field parallel to the PD, or a tensile strain is applied parallel to the PD with magnetic field perpendicular to the PD. Unlike the previously reported works on rigid exchange biased systems, a drastic decrease in exchange bias field was observed under a compressive strain with magnetic field parallel to the PD, but only a slight decrease was shown under a tensile strain. Based on a modified Stoner–Wohlfarth model calculation, we suggest that the distributions of both ferromagnetic and antiferromagnetic anisotropies are the keys to induce a mechanically tunable exchange bias.

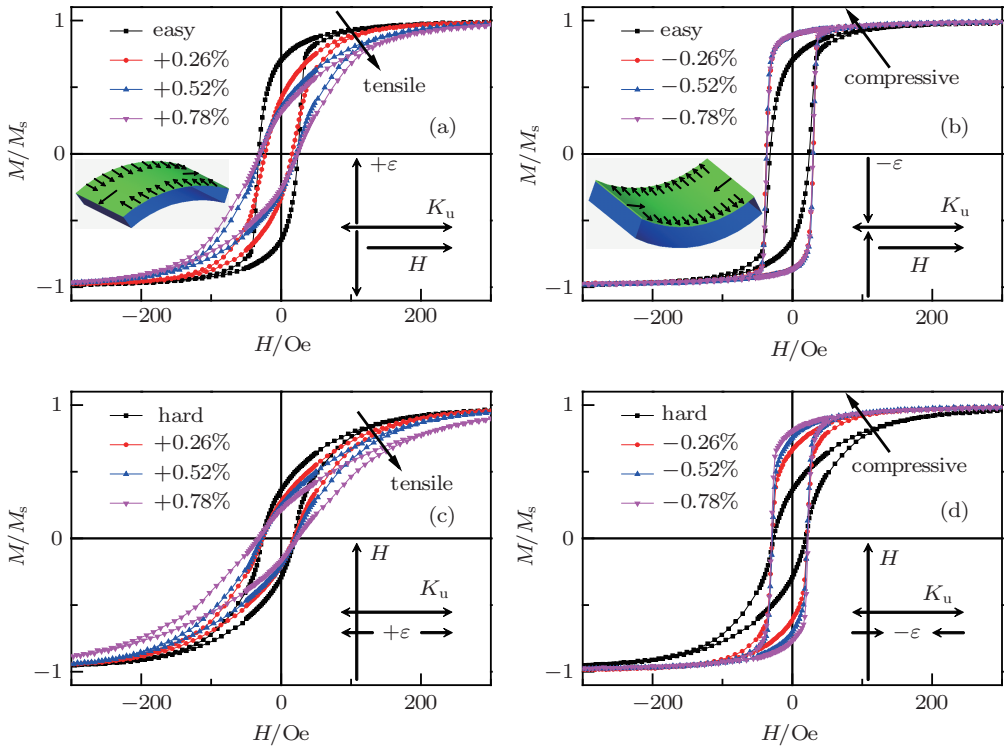


Fig. 11. Hysteresis loops for $Fe_{81}Ga_{19}$/PET obtained under various external strains using different measuring configurations: (a) magnetic field H parallel to the uniaxial anisotropy K_u and a tensile strain ε (outward bending of PET substrates) applied perpendicular to K_u, (b) H parallel to K_u and a compressive strain $-\varepsilon$ (inward bending) perpendicular to K_u, (c) H perpendicular to K_u and ε parallel to K_u, and (d) H perpendicular to K_u and ε parallel to K_u.[28]

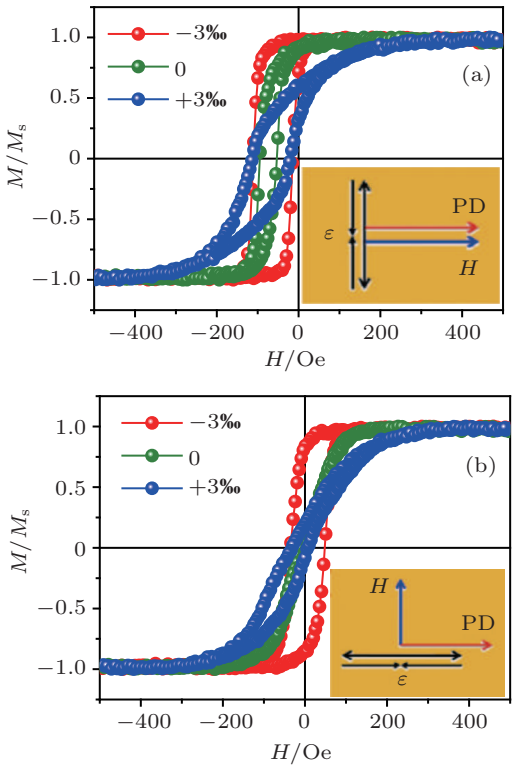


Fig. 12. The strain dependence of magnetic hysteresis loops for $Fe_{81}Ga_{19}$(10 nm)/IrMn(20 nm) bilayers with magnetic field (a) parallel and (b) perpendicular to the PD. The compressive and tensile strains are applied perpendicular or parallel to the PD, as shown in the insets of panels (a) and (b), respectively.[57]

In order to understand the effect of strain on magnetic properties microcosmically, it is necessary to image the magnetic domain structures under different strains. Chen *et al.* have prepared 100-nm Co films on polyester substrates. The magnetic domain structures with and without plastic strains were obtained by Kerr microscopy, as shown in Fig. 13.[58] It is found that the size of magnetic domains becomes much larger and ordered when the magnetic field is aligned along the easy axis. A tensile strain applied along easy axis can further increase the size of the domains. When the magnetic field is applied along the hard axis, the density of magnetic domains increases when a tensile strain is applied along the easy axis.

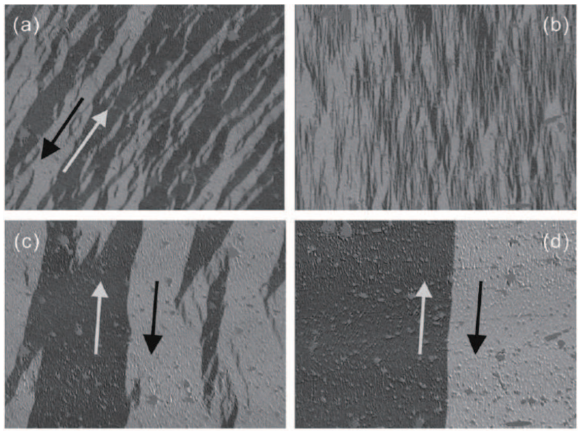


Fig. 13. Effect of strain on the magnetic domain structures of 100-nm Co films on polyester substrates. (a) Strain $\varepsilon = 0$ and H along hard axis, (b) tensile strain $\varepsilon = 0.75\%$ along easy axis and H along hard axis, (c) strain $\varepsilon = 0$ and H along easy axis, (d) tensile strain $\varepsilon = 0.75\%$ along easy axis and H along easy axis.[58]

3.3. Strain dependence of magnetoresistance

Magnetoresistance (MR), where the resistance of the material changes with applied magnetic field, has been extensively used as magnetic field sensors, read heads, and magnetic random access memory.[19] As early as 1992, Parkin *et al.* prepared flexible GMR multilayers on Kapton substrates, which display 38% room-temperature GMR, almost as large as that found in similar structures prepared on silicon wafers. Such flexible structures suggest potential technological applications in light-weight read heads.[59] In 1996, Parkin also demonstrated flexible exchange-biased magnetic sandwiches with lower saturation field and 3% room-temperature MR suggesting the possibility of manufacturing flexible MR read heads.[30] In 2010, Barraud *et al.* successfully prepared flexible Co/Al_2O_3/Co TMR structures on polyester-based organic substrates with 12.5% room-temperature TMR ratio.[60] Since the flexible substrates are easy to distort, due to the magnetostriction effect, the magnetic and transport properties of flexible magnetic films and devices strongly depend on the strain status of samples. Making the magnetic films and devices insensitive to the strain is a challenge. Melzer *et al.* demonstrated an easy approach to fabricate highly elastic spin-valve sensors insensitive to the strain on flexible PDMS.[33] By means of a predetermined periodic fracturing mechanism and random wrinkling, meander-like self-patterning devices can be achieved, as seen in Fig. 2. This meander-like structure makes the device insensitive to the strain. The influence of strain on GMR and sample resistance is shown in Fig. 14.[33] It can be clearly seen that both GMR magnitude and sample resistance maintain their values very stably under different strains, which makes them suitable to serve as stretchable sensors to detect magnetic field.

On the other hand, if MR devices are designed to evaluate strain or controlled by strain, the MR of devices is required to be sensitive to the strain. Such strain-tunable MR devices can be used in novel straintronic devices that consume extremely low power. Generally, a GMR film consists of periods of two ferromagnetic layers separated by a conducting layer, and the MR depends on the relative magnetization directions of the adjacent ferromagnetic layers and the interlayer exchange coupling.[61] An in-plane tensile strain can reduce the thickness of the spacer layers in a GMR structure, resulting in the change of interlayer exchange coupling and the MR, as Chen *et al.* have reported.[31] Another approach for strain-control of MR is to tune the magnetic anisotropy of ferromagnetic layers. As mentioned in Subsection 3.2, the magnetic anisotropy of magnetostrictive materials can be manipulated by mechanical strain or stress. Since most ferromagnetic materials exhibit the magnetostriction effect, the mechanical strain control of MR can be realized through the strain-induced magnetic anisotropy.[62,63] Özkaya *et al.* have prepared Co(8 nm)/Cu(4.2 nm)/Ni(8 nm) pseudo-spin-valve (PSV) structures on flexible PI substrates.[64] The low GMR magnitude for the as-prepared sample at zero-field indicates that the magnetization directions of Co and Ni are parallel at the remanent state, as shown in Fig. 15.[64] Applying a uniaxial strain may cause the magnetization directions in the two magnetic layers to rotate in opposite directions due to different signs of the magnetostriction coefficients of Co and Ni.

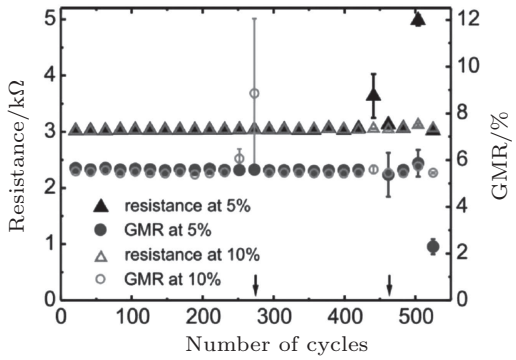


Fig. 14. GMR magnitude (circles) and sample resistance (triangles) during a cyclic loading experiment between 5% (filled symbols) and 10% (open symbols) strain.[33]

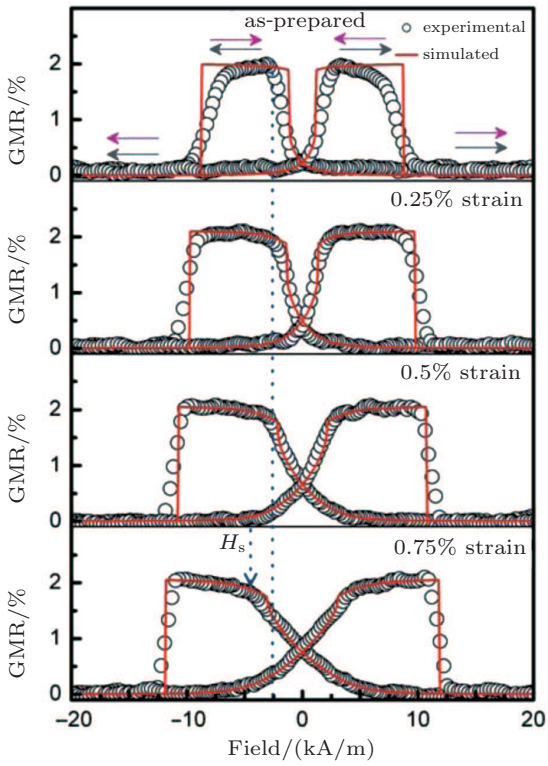


Fig. 15. GMR magnitude for Co(8 nm)/Cu(4.2 nm)/Ni(8 nm) GMR structures in (a) as-prepared state, (b) 0.25%, (c) 0.5%, and (d) 0.75% strained states.[64]

Upon applying a strain perpendicular to the easy axis of Co and Ni, the zero-field GMR magnitude increases with increasing strain, as shown in Figs. 15(b), 15(c), and 15(d), which indicates that the angle between the magnetizations of each layer in the remanent state increases. This result suggests that both the magnetic field sensitivity and the magnetic field operating range of GMR devices can be optimized by applying strain.

4. Applications

4.1. Flexible spintronic devices applied in biomedical techniques

Magnetic particles can be used to deliver drugs or genes, help to detect proteins, nucleic acids or to enhance magnetic resonance imaging (MRI) contrast, all of which are very important for modern biomedical techniques.[65–67] In a biomedical system, monitoring and analyzing the signals from magnetic particles is the essential issue, which increases the demand for integration of magnetic field sensing devices into biomedical systems. In this respect, spintronic devices, such as GMR and TMR sensors, provide an efficient solution for detecting magnetic particles due to their high magnetic field sensitivity.[68,69] However, magnetic particles usually flow in the micro-fluidic channels, so integrating the magnetic sensors with micro-fluidic channels can significantly improve their sensitivity for detecting magnetic particles. Mönch *et al.* have successfully fabricated a fully integrative rolled-up GMR sensor that simultaneously acts as a fluidic channel for in-flow detection of magnetic particles, as shown in Fig. 16.[70] This flexible and rolled-up GMR sensor leads to better signal-to-noise ratio and magnetic particles in a fluidic channel can be easily detected and counted. A small disadvantage of this rolled-up GMR sensor is that it requires intensive lithography processing, so its fabrication is expensive and time consuming. Melzer *et al.* provide another way to prepare low-cost flexible GMR sensors for use in detecting magnetic particles. The preparation process is shown in Fig. 2.[33] After preparation, the optimized GMR sensors are wrapped around the circumference of a Teflon tube, as shown in Fig. 17(a).[71] Figures 17(b) and 17(c) demonstrate the sensor's output, when the magnetic particles are passing through the flexible GMR sensors.[71]

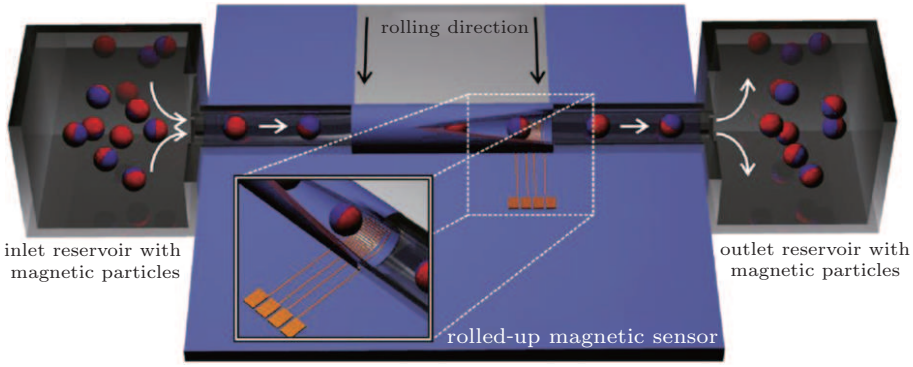


Fig. 16. Schematics revealing the main concept of rolled-up magnetic sensor for in-flow detection of magnetic particles.[70]

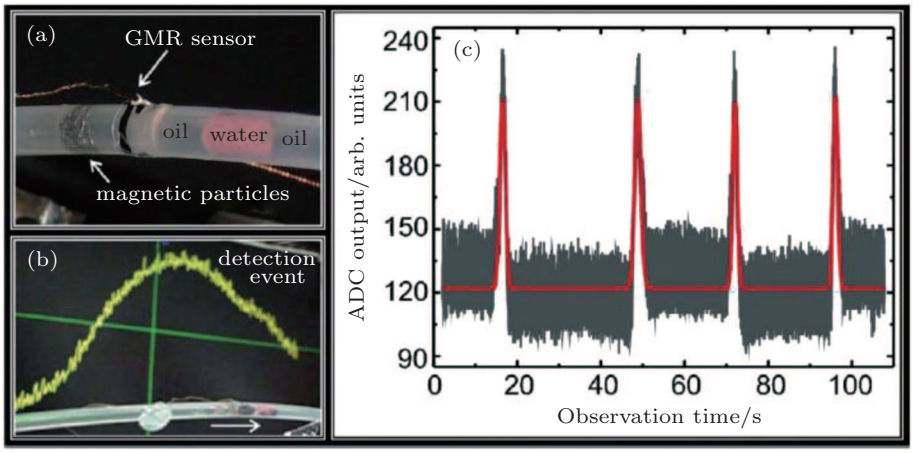


Fig. 17. Detection of magnetic particles in a fluidic channel: (a) elastic GMR sensor wrapped around the circumference of a Teflon tube. The magnetic particles are approaching the GMR sensor. (b) Signal of the elastic GMR sensor on a screen (background) as the magnetic cluster is passing the sensor (foreground). (c) Several consecutive detection events of particles passing the elastic GMR sensor.[71]

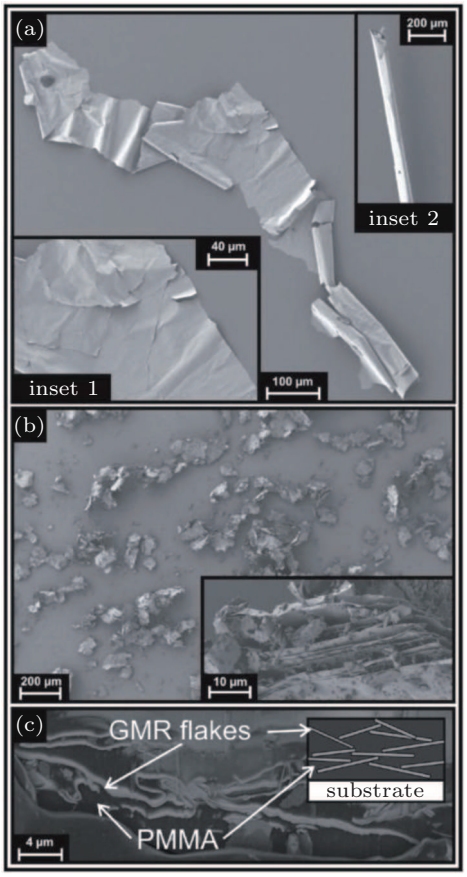


Fig. 18. SEM images of GMR powder at various stages of ink preparation: (a) initial GMR powder directly after delamination from Si substrates that consists of large metallic flakes (inset 1) and a variety of tube like structures (inset 2) self-assembled by releasing the film's intrinsic stress. (b) The magnetic film is milled using ceramic beads in order to produce magnetic powder consisted of variously shaped flakes. (c) SEM image of a cross section of a printed sensor shows the internal structure of metallic flakes percolated inside polymer; (inset) schematic drawing illustrates the principle of flake percolation.[73]

The flexible electronic circuits used in biomedical systems are generally prepared by means of a newly developed printing method that could revolutionize large-area and low-cost electronics manufacturing.[72] However, the fabrication of printable magnetic sensors remains challenging due to a lack of magnetic inks containing various components. Karnaushenko *et al.* have for the first time developed a kind of magnetic ink with GMR flakes which can be easily printed on various substrates, such as paper, polymer, and ceramic.[73] In order to fabricate the magnetic ink, GMR sensors are first deposited on 3-inch silicon wafers with photosensitive polymer AR-P 3510 as the buffer layer. After preparation, the samples are rinsed in acetone to release the deposited GMR sensors from the substrates. The obtained GMR sensors show flake-like or rolled-up structures arising from the intrinsic strain of the deposited GMR films on rigid substrates, as shown in Fig. 18(a).[73] To assure high electrical conductivity of as-prepared GMR flakes, a multilayer stacked structure is prepared from the originally obtained material by ball milling. The resulting powder is filtered through a grid that defines the maximum lateral size of a GMR flake to about 150 μm, as shown in Fig. 18(b). A GMR ink is prepared by mixing 500 mg of the GMR powder with 1 ml of a binder solution that is an acrylic rubber based on poly (methyl methacrylate) (PMMA) dissolved in a methyl isobutyl ketone. Finally, using a brush, the solution is painted on different surfaces, i.e. paper, polymers, and ceramic. In Fig. 18(c), a cross-section scanning electron microscopy (SEM) image of a large-area film shows continuous layer-stacked structures, which significantly facilitate the electron transport along the in-plane direction to achieve a high GMR effect. This method uses standard sputter deposition, milling, and mixing machines for high yield production, demonstrating the suitability of the printable magnetoelectronic devices for large scale industrial production.

4.2. Flexible multiferroic structure applied in energy harvesting

Energy harvesting is a process by which energy can be captured from external sources, such as solar power, thermal energy, vibrational energy, electromagnetic waves, etc., and converted into electrical energy.[74] Energy harvesting technologies can be substituted for batteries, minimizing power consumption. In particular, there is significant interest in harvesting vibration energy by using piezoelectric and magnetic devices.[75,76] Figure 19 shows a magnetoelectric energy harvesting mechanism of multiferroic composites made by combining magnetostrictive and piezoelectric phases together.[39] First, an external magnetic field leads to deformation of the magnetostrictive phase through the magnetostriction effect, and the deformation can be transmitted to the piezoelectric phase across the interface between the two phases, generating electrical charges due to the converse piezoelectric effect. With the development of wearable electronics, more and more electronics are increasingly integrated into clothing, either for functional or fashion reasons. This requires the next generation of energy harvesters to move into wearable electronics, which needs the magnetic and piezoelectric materials to be flexible and lightweight. Onuta *et al.* have fabricated an electromagnetic energy harvester consisting of a magnetostrictive $Fe_{0.7}Ga_{0.3}$ thin film and a $Pb(Zr_{0.52}Ti_{0.48})O_3$ piezoelectric thin film on a 3.8-μm-thick Si cantilever, as shown in Fig. 4.[38] The dependence of output voltage and harvested power on the AC magnetic field is shown in Fig. 20.[38] The peak harvested power of 0.7 mW/cm^3 at 1 Oe is about 6 times larger than the value reported in Terfenol-D/PZT/Terfenol-D laminated structures,[77,78] which promotes the development of flexible energy harvesting materials.

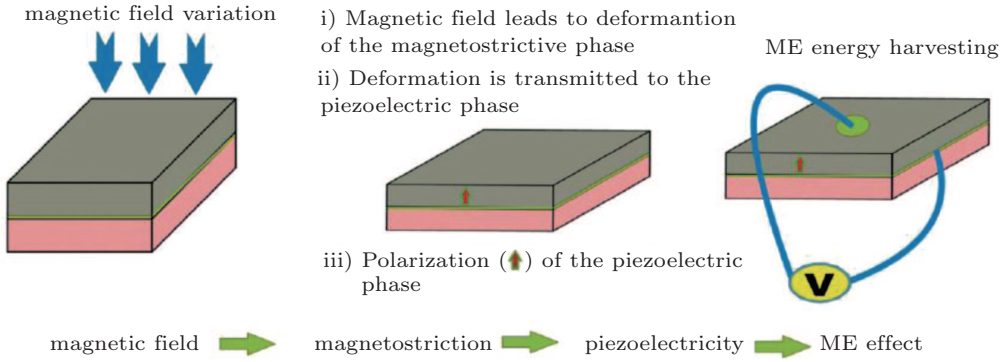


Fig. 19. Schematic show of harvesting mechanism for multiferroic materials.[39]

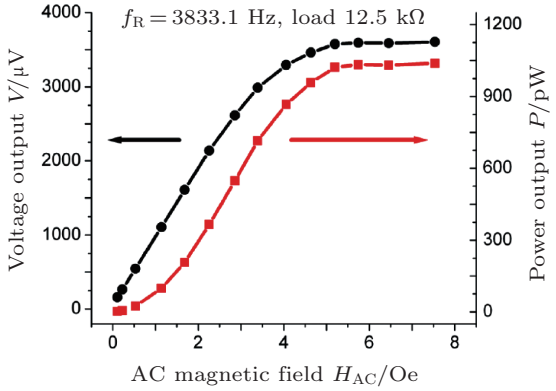


Fig. 20. Dependence of the output voltage and harvested power on the AC magnetic field of $Pb(Zr_{0.52}Ti_{0.48})O_3$/ $Fe_{0.7}Ga_{0.3}$ cantilevers.[38]

4.3. Flexible polymer-based magnetic composites applied in actuators

Polymer-based magnetic composites are flexible, lightweight, and easily processed, and they are widely applied in MEMS.[79] The magnetically actuated micro-devices can be controlled without wire as long as the actuation environment is magnetically transparent, and therefore can be operated in air, vacuum, water, etc.[80] The flexible, wireless control of micro-devices makes polymer-based magnetic composite attractive for many applications. However, the actuation and precise control of the polymeric components is a difficult issue. Lee *et al.* have prepared an actuator consisting of a body plate 50-μm thick and four legs using photoresist SU-8.[81] Three square sections of NiFe film 10-μm thick are in turn electroplated on the top and bottom sides of the SU-8 body, as shown in Fig. 21.[81] When the magnetic field is applied to the actuator along the longitudinal direction, the magnetostriction of NiFe films makes the actuator curve and move along the field direction, which is useful in micro-machines area such as magnetic field driven drug delivery. Kim *et al.* have presented a new magnetic polymeric micro-actuator that allows the programming of heterogeneous magnetic anisotropy at microscale.[82]

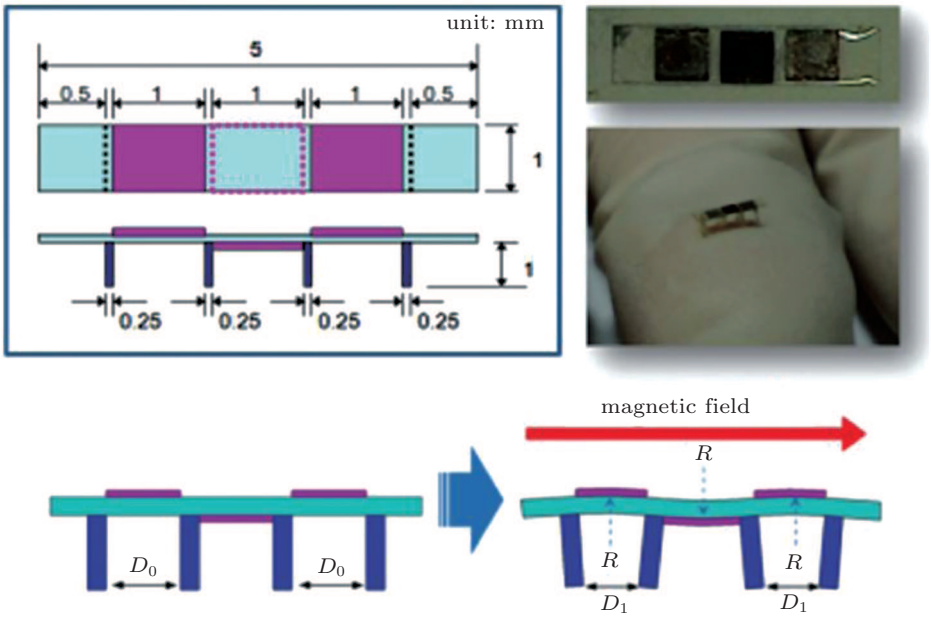


Fig. 21. Schematic view with photograph of a fabricated NiFe thin film worm actuator and its deflected motion.[81]

As seen in Fig. 22, the micro-actuator is composed of four magnetic bodies having different magnetic easy axes, such that it has various configurations according to the applied external magnetic field direction. By freely programming the rotational axis of each component, the polymeric micro-actuators can undergo predesigned, complex two- and three-dimensional motions.

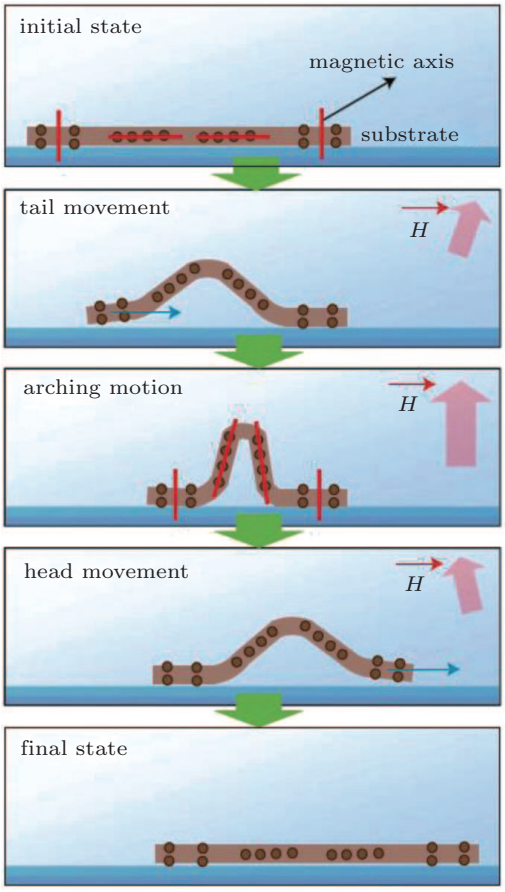


Fig. 22. Movement of polymeric micro-actuators by magnetic field.[82]

4.4. Flexible soft magnetic films applied in microwave devices

Flexible soft magnetic films exhibit high microwave permeability, which is of practical importance for a number of applications, such as high frequency inductors, transformers, shielding, and electromagnetic interference (EMI) devices.[83–85] Additionally, the flexible films can be cut easily and be used on many different surfaces. Due to the magnetostriction effect, the magnetic anisotropy of flexible soft magnetic films can be tuned by changing the status of strain in flexible films, which provides the possibility to overcome the Snoek limit and design frequency-tunable microwave devices.[86,87] Flexible thin films of magnetic alloys, such as FeCoB, FeCoBSi, FeTaN, FeZrN, and CoAlO, and polymer-nanoparticle composites have been prepared due to their high frequency applications.[88–93] Zuo *et al.* have prepared [Fe–Co–Si (d)/native oxide]$_{50}$ multilayer films with different metallic layer thicknesses (d) on flexible Kapton substrates by DC magnetron sputtering.[94] Figure 23 depicts the permeability spectra for the multilayer films with various d values. The films exhibit relatively high complex permeability and ferromagnetic resonance frequencies up to 7.9 GHz, indicating great potential for applications in high-frequency electromagnetic devices. Rasoanoavy *et al.* have prepared a flexible CoFeB/PVDF/CoFeB composite material and observed a 30% variation in its microwave permeability under a 1.5-MV/m electric field, due to the modified magnetic anisotropy caused by the strain-mediated magnetoelectric effect.[95]

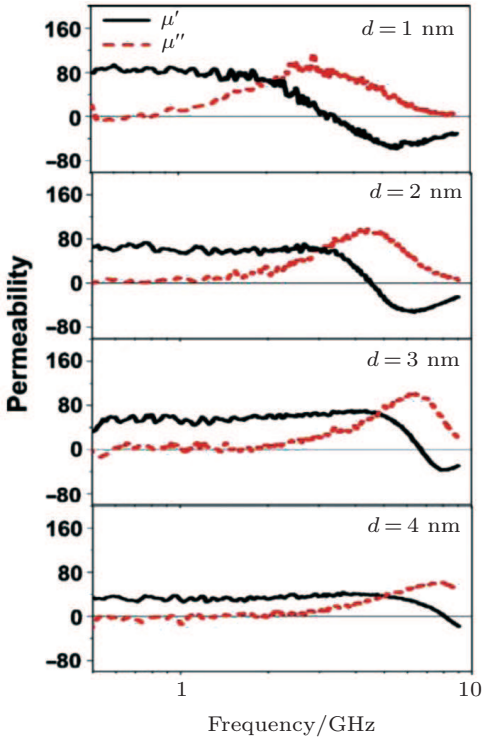


Fig. 23. Permeability spectra of [Fe–Co–Si (d)/native oxide]$_{50}$ multilayer films.[94]

Employing the exchange bias effect is another way to promote the resonance frequency of the magnetic films and devices. Phuoc *et al.* have prepared permalloy–FeMn multilayers on flexible Kapton substrates.[96] A multiple-stage magnetization reversal and consequent plural ferromagnetic resonance absorption was observed, which may be due to the different exchange interfacial energies acting on each layer. Based on these results, they have demonstrated a wide-band microwave absorber by using flexible permalloy–FeMn multilayers. Figure 24 shows the frequency dependence of the reflection loss of flexible permalloy–FeMn multilayers. The working bandwidth (the absorption width where the reflection loss is less than 10 dB) of the present film is rather broad, ranging from 1 GHz to 4 GHz, indicating that flexible

exchange-biased multilayer systems are promising for future high-frequency applications.

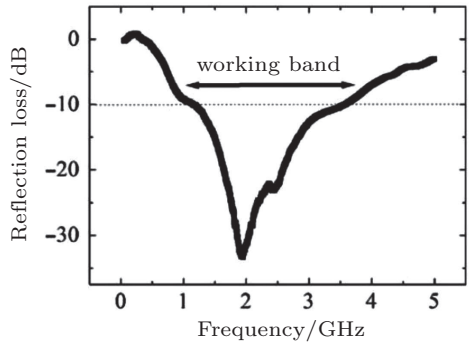


Fig. 24. Frequency dependence of the reflection loss of flexible permalloy–FeMn multilayers.[96]

5. Conclusions and perspectives

The investigation of flexible magnetic films and their applications is an intense new trend, which is potentially important for the development of flexible electronics. In order to foster a comprehensive understanding of the flexible magnetic films and related applications, we have reviewed recent advances in the study of flexible magnetic films, including the fabrication methods, the physical properties, and the applications of the films and devices. (i) One can prepare the desired flexible magnetic films by deposition of magnetic films on flexible substrates, by a transfer and bonding approach or by including and then removing sacrificial layers. Generally, all flexible magnetic films require fabrication on a supporting flexible substrate. Therefore, flexible substrates with good flatness, high treatment temperature, good thermal stability, good mechanical properties, etc., are very important for the fabrication of high quality flexible magnetic films. (ii) Due to the flexibility of flexible magnetic films, the strain effect on the magnetic-related properties is very important, both for fundamental research and for practical applications. The magnetic anisotropy, exchange bias, coercivity, and magnetoresistance of flexible magnetic films and devices can be effectively manipulated by externally applying strain, which has good potential for applications in straintronic devices. However, in order to realize multifunctional straintronic devices, further comprehensive studies on the physical properties of flexible magnetic films are necessary, especially under multi-field conditions, like electric field, magnetic field, strain field, etc. (iii) Flexible magnetic films can be applied in the spintronic devices, energy harvesters, actuators, microwave devices, etc., due to the properties of bio-compatible, low-cost, light-weight, and compact.

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