Magnetostrictions and Magnetic Properties of Nd-Fe-B and $SrFe_{12}O_{19}$ *

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The magnetostrictions of polycrystalline Nd-Fe-B and Sr-ferrite at different temperatures are reinvestigated using a strain gauge rotating-sample method. It is found that the magnetostriction λ_s of Nd-Fe-B is $+52\times10^{-6}$, and that of Sr-ferrite is -25×10^{-6} under a magnetic field of 8 T at room temperature. The maximum energy product $(BH)_{\max}$ of the Nd-Fe-B magnet is improved when the powders are magnetically aligned perpendicular to the pressing direction, whereas that of the Sr-ferrite magnet is better when the powders are aligned parallel to the pressing direction. These experimental results suggest that the magnetostriction can generate compressive strain anisotropy resulting from the inverse effect of the magnetostriction. Thus, the magnetization of materials with a negative coefficient of magnetostriction are easier to be aligned normal to the stress direction, while for the materials with a positive coefficient of magnetostriction, the magnetization is easier to be aligned along the stress direction. Therefore, the magnetostriction anisotropy can be used to improve the alignment of the magnetic powders as well as the performance of the magnets.

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Nd-Fe-B and hard ferrites are the most important permanent magnetic materials for commercial applications. Hard ferrites such as Ba-ferrite ($BaFe_{12}O_{19}$) and Sr-ferrite ($SrFe_{12}O_{19}$) were developed in the 1950s and have been widely used in modern electronics due to their low costs. The Nd-Fe-B ($Nd_2Fe_{14}B$) magnetic material was developed in 1983 and its production and applications have increased enormously in the past twenty years because of its outstanding magnetic properties around room temperature. Both Nd-Fe-B and the hard ferrites have been extensively studied,[1−6] however, only a few groups have reported their magnetostrictions. Mgnetostriction is one of the most important factors in determining the magnetic properties of hard magnetic materials. Using a traditional strain gauge method, two groups studied the magnetostrictions of $Nd_2Fe_{14}B$, but discrepant results were obtained.[7−9] The work by Graham *et al.*[7] was based on aligned sintered samples. Their results indicated that the magnetostriction of $Nd_2Fe_{14}B$ is relatively small ($+10$ to $+20\times10^{-6}$ at low field, parallel or perpendicular to the magnetic field). However, Ibarra *et al.*[8,9] measured the polycrystalline Nd-Fe-B sample under a plus magnetic field and reported a magnetostriction of $\lambda_t = -96\times10^{-6}$ ($\lambda_t = \lambda_{\parallel} - \lambda_{\perp}$) at room temperature. The longitudinal magnetostriction of $SrFe_{12}O_{19}$ as a function of magnetic field was investigated by Grössinger *et al.*[10] using a miniature capacitance dilatometer at RT. The longitudinal magnetostriction is about -2×10^{-5} under a magnetic field of 8 T at RT.

In this study, the strain gauge rotating-sample method is used to measure the magnetostrictions of Nd-Fe-B and $SrFe_{12}O_{19}$. It is found that the magnetostriction λ_s of Nd-Fe-B is positive at room temperature, while that of $SrFe_{12}O_{19}$ is negative. The maximum energy product $(BH)_{\max}$ of Nd-Fe-B magnets is improved when the sample powders are pressed in the direction perpendicular to the aligning magnetic field whereas that of $SrFe_{12}O_{19}$ magnets is better when pressed along the direction parallel to the applied field. It is possible that this disparity is closely associated with the different signs of their magnetostriction.

The Nd-Fe-B powders were produced using a hydrogenation disproportionation desorption recombination (HDDR) process. The Sr-ferrite powders were purchased from MGRIMM magnetic material technology Co., Ltd. Both Nd-Fe-B and $SrFe_{12}O_{19}$ powders are anisotropic. For magnetostriction measurements, the powder samples were bonded with epoxy resin with a volume fraction of ∼15% for Nd-Fe-B, (∼17% for Sr-ferrite), to ensure the isotropic bonded state of the samples. The measurement results were modified according to Ref. [11]. The effective magnetostriction

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ratio λ^* is defined as

$$\lambda^* \cong \frac{f}{1-1.25\sigma_2}(1.1538-1.2628\sigma_2-0.2122f^{2/3}), \quad (1)$$

$$\lambda \quad = \quad \lambda(f)/\lambda^*, \quad (2)$$

where f is the filling volume fraction of the magnetostriction component; σ_2 is the Poisson ratio of the nonmagnetostriction medium (σ_2=0.4 for epoxy resin). Thus, the λ^* values are obtained to be about 0.18 and 0.2 for Nd-Fe-B and $SrFe_{12}O_{19}$, respectively. The bonded samples are in dimensions $10\times8\times1\,\mathrm{mm}^3$.

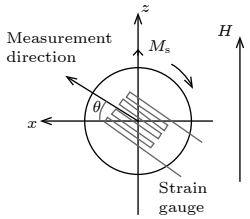

Fig. 1. Illustration of the rotating-sample measurement method.

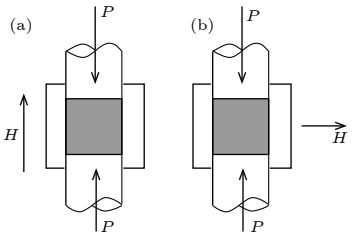

Fig. 2. Different magnetic alignment processes for magnetic powders: (a) axial die-pressing $P \parallel H$ and (b) transverse die-pressing $P \perp H$.

The magnetostriction measurements were carried out using a Wheatstone bridge. The strain gauges were bonded to the surface of the test samples. To minimize the magnetoresistance and thermal expansion effects on the strain gauge, a compensation strain gauge in the bridge circuit was glued on a copper plate and was placed together with the measured samples. The magnetostrictions as a function of angle were measured using the horizontal sample rotator option of a physical property measurement system (PPMS, Quantum Design Inc.), which can turn the sample from 0° to 360° in the magnetic field. The sample plane is always perpendicular to the rotating axis. Three wires from the measurement strain gauge and compensation strain gauge were connected to the strain bridge. A magnetic field of up to 8 T was applied during the measurements. Figure 1 illustrates the rotating-sample measurement method. The starting angle of the rotator (θ=0) corresponds to the $\lambda_\perp$ direction. The strain bridge was tuned to the balance state in a magnetic field of 8 T before rotating measurements, i.e. the reading data on the bridge correspond to $(\lambda(\theta)-\lambda_\perp)$. Here we refer it as the rotating-sample method. We also measured the magnetostriction $(\lambda_\parallel - \lambda_\perp)$ as a function of the applied magnetic field H. In this study, we refer to this method as the conventional method. The data acquisition and processing were performed by a computer using the IEEE-488 interface bus and the Labview software.

In order to study the effect of the magnetostriction on the magnetic properties of these materials, magnets of sintered Nd-Fe-B and bonded Sr-ferrite have been prepared using the magnetic powders with different magnetic aligning processes as shown in Fig. 2. A magnetic field of 2 T was applied along the direction parallel or perpendicular to the pressing direction.

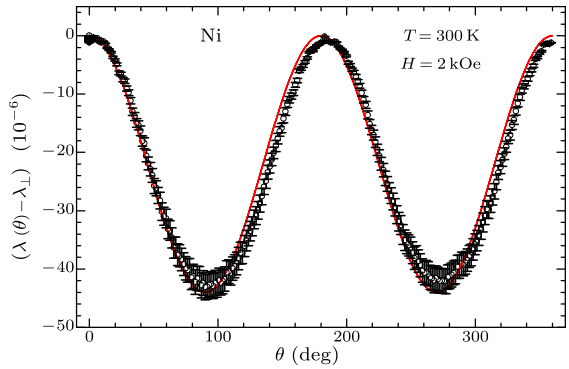

Fig. 3. The magnetostriction $\lambda(\theta)-\lambda_\perp$ of Ni at 300 K as a function of measurement angle θ (H=2 kOe). Open circle: experimental results. Solid line: calculated results.

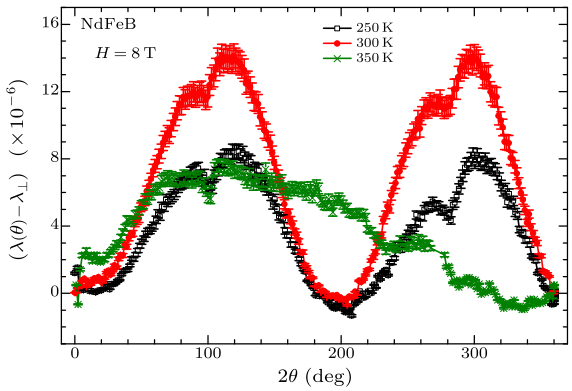

Fig. 4. The magnetostriction $\lambda(\theta)-\lambda_\perp$ of Nd-Fe-B as a function of measurement angle θ at different temperatures (H=8 T).

In order to test whether the magnetostriction $[\lambda(\theta)-\lambda_\perp]$ can be accurately measured using the rotating-sample method, the magnetostriction of a polycrystalline Ni plate-like sample was measured as a function of the measured angle θ (Fig. 3). The open dots are corresponding to the experimental data and the solid line represents the calculated results using the formula

$$\lambda(\theta) = \frac{3}{2}\overline{\lambda}\Big(\cos^2\varphi - \frac{1}{3}\Big). \quad (3)$$

It should be pointed out that $\overline{\lambda} = \frac{3}{2}(\lambda_\parallel - \lambda_\perp)$ for the isotropic polycrystalline magnetic materials, where ϕ

is the angle between the magnetization and the measurement direction (note: $\phi = 90° - \theta$ since the starting angle of the rotator (θ=0) corresponds to the $\lambda_\perp$ direction). The $\overline{\lambda}$ is the saturation magnetostriction for polycrystalline materials measured along the magnetic field direction. As shown in Fig. 3, the calculated results agree very well with the measured data. According to this figure and the Eq. (3), we can obtain that $(\lambda_\parallel - \lambda_\perp) = -45 \times 10^{-6}$, $\lambda_s = -30 \times 10^{-6}$, and $\lambda_\perp = +15 \times 10^{-6}$, which are in good agreement with the standard value of Ni ($\lambda_s = -33 \times 10^{-6}$). Thus we believe that the rotating-sample method is accurate enough to investigate the magnetostriction of magnetic materials with a similar magnitude of magnetostriction.

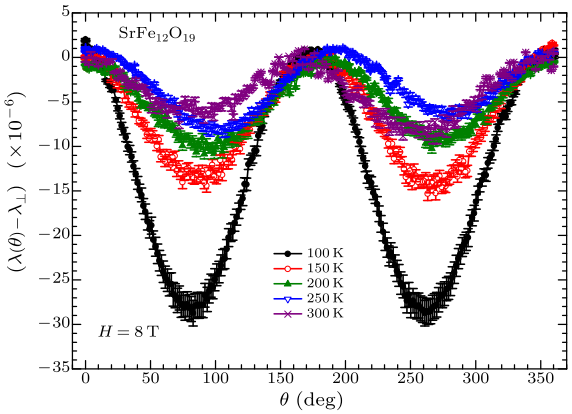

Fig. 5. The magnetostriction $\lambda(\theta) - \lambda_\perp$ of $SrFe_{12}O_{19}$ as a function of measurement angle θ at different temperatures (H=8 T).

The rotating-sample method was then applied to Nd-Fe-B. Figure 4 shows the $(\lambda(\theta) - \lambda_\perp)$ vs θ curves of Nd-Fe-B measured under a magnetic field of 8 T at temperatures of 250, 300 and 350 K, respectively. It was found that the magnetostriction λ_s of Nd-Fe-B is about $+52 \times 10^{-6}$ at room temperature. This result is different from that of Ibarra *et al.*,[8] but similar to the data of (IIb–IIa) by Graham *et al.*[7] Two peaks shown around 90° and 270° may be attributed to the preferred orientation of the 180° domains in the Nd-Fe-B sample. The $(\lambda(\theta) - \lambda_\perp)$ curves of Nd-Fe-B at 250 and 300 K show the shape of cosine functions (Fig. 4). The unexpected curve at 350 K may result from the strain gauge or the binder failure at high temperature. The curves of $(\lambda(\theta) - \lambda_\perp)$ vs θ for Nd-Fe-B under a magnetic field of 8 T at T $\leq$ 200 K do not also follow the shape of cosine function (data not shown), probably due to the increase of magnetocrystalline anisotropy field at low temperature. The anisotropy field μH_a of $Nd_2Fe_{14}B$ is about 7.5 T at room temperature and increases monotonically with the decreasing temperature even though spin reorientation occurs at about 135 K.[12]

The curves of $\lambda(\theta) - \lambda_\perp$ vs θ of $SrFe_{12}O_{19}$ measured under a magnetic field of 8 T at different temperatures are shown in Fig. 5. The magnetostriction of $SrFe_{12}O_{19}$ is negative, and the values of magneostriction $\lambda_s = -95, -49, -33, -28, -25 \times 10^{-6}$ at 100, 150, 200, 250, and 300 K respectively. The value of magnetostriction at RT is in good agreement with that obtained by Grössinger *et al.*[10] The curves at different temperatures show the typical shape of cosine functions (Fig. 5).

Table 1. The magnetic properties of magnets made with different aligning processes. Here B_r is the remanence. The data in parentheses are in units of electromagnetism.

Magnetic material	Press manner	B_r (T or kG)	H_{cj} (MA·m^{-1} or kOe)	$(BH)_{max}$ (kJ·m^{-3} or MGOe)
Nd-Fe-B (sintered)	$\parallel H$	1.13 (11.3)	1.66 (20.8)	239.2 (30.1)
	$\perp H$	1.21 (12.1)	1.65 (20.7)	280.8 (35.3)
Sr-ferrite (bonded)	$\parallel H$	0.276 (2.76)	0.164 (2.06)	13.0 (1.65)
	$\perp H$	0.234 (2.34)	0.156 (1.96)	9.0 (1.15)

The magnetic properties of Nd-Fe-B and $SrFe_{12}O_{19}$ bonded magnets made with different magnetic aligning processes are listed in Table 1. For Nd-Fe-B sintered magnets, the magnet aligned perpendicular to the pressing direction shows a higher maximum energy product $(BH)_{max}$, whereas for $SrFe_{12}O_{19}$ bonded magnets, the sample aligned parallel to the pressing direction has a better performance. Compared to the samples aligned parallel to the pressing direction, the magnetic remanence of perpendicularly aligned Nd-Fe-B sample increases by about 0.8 kG and the maximum energy product $(BH)_{max}$ increases by 5.2 MGOe. The latter is 17% higher than the sample aligned parallel. Whereas the magnetic remanence of the $SrFe_{12}O_{19}$ sample aligned parallel to the pressing direction increases 0.4 kG and the $(BH)_{max}$ increases 0.5 MGOe (43% higher) compared to that of the samples aligned perpendicular to the pressing direction. In order to understand these phenomena, the inverse magnetostriction effect (the magnetomechanical effect) was considered. The magnetoelastic energy is given by[13]

$$E_{me} = -\frac{3}{2}\lambda\sigma\cos^2\varphi, \tag{4}$$

where σ is the applied stress, λ is the magnetostriction

constant and φ is the angle between M_s and the stress direction. The compressive stress is negative ($\sigma < 0$) during the magnetic alignment. Therefore, if $\lambda < 0$, the energy will be minimal when the magnetization (or magnetic field) is parallel to the stress direction (i.e. ϕ=0). If $\lambda > 0$, the energy will be minimal when the magnetization (or magnetic field) is perpendicular to the stress direction (i.e. $\phi = 90°$). In other words, a compressive stress σ will help to orient magnetic powders parallel to the direction of the applied magnetic field H, when $H \parallel \sigma$ for $\lambda < 0$ materials, or $H \perp \sigma$ for $\lambda > 0$ materials due to the magnetoelastic energy. Therefore, the magnetostriction is likely to be considered as one of the main factors to improve the alignment of magnetic powders. According to the negative or positive magnetostriction of the magnetic materials, different pressing directions should be chosen.

In summary, we have found that the magnetostriction of $Nd_2Fe_{14}B$ is positive ($\lambda_s = +52 \times 10^{-6}$) at room temperature, whereas the magnetostriction of $SrFe_{12}O_{19}$ is negative ($\lambda_s = -95 \times 10^{-6} - -24 \times 10^{-6}$) at temperatures ranging from 100 to 300 K. The experimental results demonstrate that the inverse effects of magnetostriction may result in compressive strain anisotropy. In order to further improve the maximum energy product $(BH)_{\max}$ of the magnets, pressing along perpendicular direction to the magnetic field is better for $\lambda > 0$ materials, while pressing along the magnetic field direction is better for $\lambda < 0$ materials. This magnetostriction anisotropy may thus be used to improve the performance of magnets in the fabrication process.

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References

[1] Goldman A 2006 *Modern Ferrite Technology* 2nd edn (Pittsburgh, PA: Springer)
[2] Sagawa M, Fujimura S, Togawa M and Matsuura Y 1984 *J. Appl. Phys.* **55** 2083
[3] Croat J J, Herbst J F, Lee R W and Pinkerton F E 1984 *J. Appl. Phys.* **55** 2078
[4] Buschow K H J 1986 *Mater. Sci. Rep.* **1** 1
[5] Herbst J F 1991 *Rev. Mod. Phys.* **63** 819
[6] Yang N, Dennis K W, McCallum R W, Kramer M J, Zhang Y and Lee P L 2005 *J. Magn. Magn. Mater.* **295** 65
[7] Graham C D Jr and Flanders P J 1986 *IEEE Trans. Magn.* **22** 749
[8] Ibarra M R , Algarabel P A, Alberdi A, Bartolomé J and del Moral A 1987 *J. Appl. Phys.* **61** 3451
[9] Algarabel P A, Ibarra M R, Marquina C and Moral A de1 1990 *J. Magn. Magn. Mater.* **84** 109
[10] Grossinger R et al 2007 *J. Magn. Magn. Mater.* **310** 2587
[11] Herbst J F, Capehart T W and Pinkerton F E 1997 *Appl. Phys. Lett.* **70** 3041
[12] Grössinger R, Sun X K, Eibler R, Buschow K H J and Kirchmayr H R 1986 *J. Magn. Magn. Mater.* **58** 55
[13] Cullity B D 1972 *Introduction to Magnetic Materials* (Reading, Mass.: Addison-Wesley) p 266