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Giant magnetostriction in $Sm_{1−x}Nd_xFe_{1.93}$ compounds

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The polycrystalline $Sm_{1−x}Nd_xFe_{1.93}$ ($x=0$, 0.08, 0.12, 0.16, 0.2, 0.24, 0.32, and 0.36) compounds are found to stabilize in $MgCu_2$-type C15 cubic Laves phase structure. The compound $x=0.12$ is found to exhibit giant magnetostriction value of -1572×10^{-6} due to the anisotropy compensation between Sm^{3+} and Nd^{3+} ions. The easy direction of magnetization (EMD) is observed towards $\langle 111 \rangle$ for the $0 \leq x \leq 0.20$ compounds, accompanied by a rhombohedral distortion. For the $x=0.32$ compound, an orthorhombic distortion, with a support that the EMD is towards $\langle 110 \rangle$, is observed. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2751124]

The binary C15 cubic Laves phase RFe_2 (R=rare-earth elements) compounds are known to exhibit giant magnetostriction values and large magnetocrystalline anisotropy at room temperature.[1,2] Polycrystalline compounds of $TbFe_2$, $DyFe_2$, and $SmFe_2$ have been reported to have the saturation magnetostriction (λ_s) of 1753×10^{-6}, 433×10^{-6}, and -1560×10^{-6}, respectively. Clark,[1] have reported $Tb_{0.27}Dy_{0.73}Fe_2$, the anisotropy compensated pseudobinary compound, that was prepared from $TbFe_2$ and $DyFe_2$ whose first order anisotropy constants (K_1) have opposite signs. The polycrystalline anisotropy compensated compounds $Tb_{0.27}Dy_{0.73}Fe_2$, $Sm_{0.86}Dy_{0.14}Fe_2$, and $Sm_{0.9}Pr_{0.1}Fe_2$ have been reported to have ($\lambda_\parallel - \lambda_\perp$) of 1500×10^{-6}, -1688×10^{-6}, and -1600×10^{-6}, respectively, at room temperature.[3–5] According to the single ion model proposed by Clark *et al.*[1] the spontaneous magnetostriction (λ_{111}) values of $NdFe_2$, $PrFe_2$, $DyFe_2$, and $SmFe_2$ are 2000×10^{-6}, 5600×10^{-6}, 4200×10^{-6}, and -3200×10^{-6}, respectively, at 0 K. However, $TbFe_2$ and $SmFe_2$ have negative K_1 values and are known to possess opposite signs of magnetostriction values. Recently, Shi *et al.*[6] reported the anisotropy compensation and magnetostriction studies on high pressure-synthesized $Tb_xNd_{1−x}Fe_{1.9}$ compounds. $PrFe_2$ and $NdFe_2$ compounds do not form in single phase through conventional preparation methods and therefore, the nature of anisotropy of these are unknown.[1] The easy magnetization direction (EMD) of the high pressure-synthesized $NdFe_2$ compound is nearly along the $\langle 100 \rangle$ direction at room temperature.[7] It has also been reported that the EMD of $SmFe_2$ is along the $\langle 111 \rangle$ direction accompanied by the rhombohedral distortion at the room temperature.[1] In this letter, we report the anisotropy compensation in $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ compound accompanied by giant negative magnetostriction at room temperature.

The $Sm_{1−x}Nd_xFe_{1.93}$ ($x=0$, 0.08, 0.12, 0.16, 0.2, 0.24, 0.32, and 0.36) compounds were prepared by arc melting the constituent elements (rare-earth elements: 99.9% pure, Fe: 99.95% pure) in an arc furnace under argon atmosphere. Each compound was melted several times for homogeneous mixing and the weight loss after the final melting was less than 0.4%. The ingots were sealed in quartz ampoules under $\sim 10^{-6}$ mbar pressure and were annealed at 600 °C for 5 days. Powder x-ray diffraction (XRD) patterns of the compounds were taken using a PANalytical (X'pert PRO) x-ray diffractometer employing Cu $K\alpha$ radiation. The compositional analyses were carried out using the energy dispersive analysis of x rays (EDAX) in scanning electron microscope. The room temperature magnetostriction measurements up to a magnetic field of 11 kOe were carried out on the rod shaped (4 mm diameter 12 mm long) samples of the compounds, employing the strain gauge method.[8] In order to determine the EMD of the above compounds, samples were prepared using the fine powders of the compounds mixed with an epoxy resin. These samples were aligned in the presence of a magnetic field of 20 kOe, and the epoxy resin was allowed to cure for 30 min. The EMDs in the compounds were determined from the presence of certain reflections and the absence of the rest in the XRD patterns of the aligned samples.

The powder XRD patterns of $Sm_{1−x}Nd_xFe_{1.93}$ ($x=0$, 0.12, 0.24, 0.32, and 0.36) compounds are shown in Fig. 1. The compounds have formed in single phase with the cubic Laves phase structure up to $x=0.32$. Small quantities of the 1:3 phase ($PuNi_3$-type crystal structure) is also observed along with the Laves phase for the $x=0.36$ compound. As $NdFe_2$ has not been stabilized through the conventional preparation technique, the Nd rich compounds may contain the 1:3 phase as an additional phase. The lattice parameter (Fig. 2) slightly increases with Nd content, in accordance
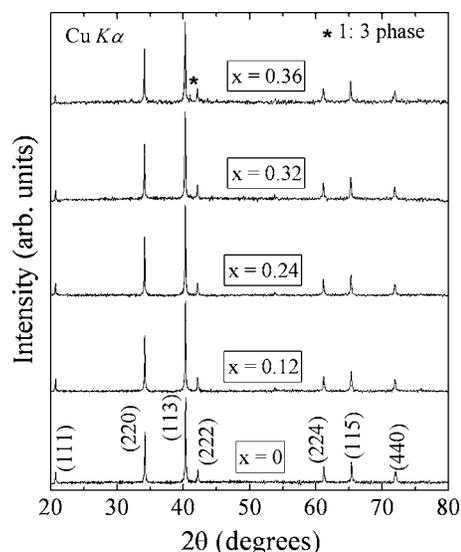

FIG. 1. XRD patterns of $Sm_{1−x}Nd_xFe_{1.93}$ compounds.

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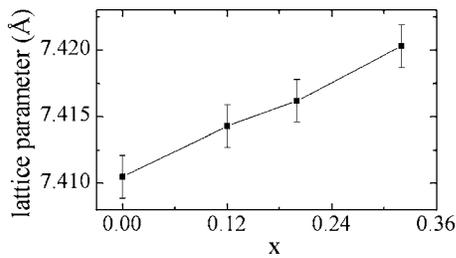FIG. 2. Variation of lattice parameter of $Sm_{1-x}Nd_xFe_{1.93}$ compounds with x.

FIG. 4. Magnetostriction curves of $Sm_{1-x}Nd_xFe_{1.93}$ compounds at room temperature.

with the larger size of Nd^{3+} ions than that of Sm^{3+} ion. The EDAX patterns of all the $Sm_{1-x}Nd_xFe_{1.93}$ ($x=0$, 0.08, 0.12, 0.16, 0.20, 0.24, 0.32, and 0.36) compounds are shown in Fig. 3 and the inset boxes represent the amounts (at. %) of the individual elements in the compounds. The intensity of the peak corresponding to Nd is found to increase with increase of the Nd content. The atomic percentages of the constituent elements in the above compounds are in agreement with the nominal compositions taken.

The room temperature magnetostriction curves of the $Sm_{1-x}Nd_xFe_{1.93}$ ($x=0$, 0.08, 0.12, 0.16, and 0.20) compounds are shown in Fig. 4. $\lambda_\parallel$ and $\lambda_\perp$ corresponds to the strain when the magnetic field is applied along and perpendicular to the axis of the rod, respectively. The $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ compound is seen to have large parallel and perpendicular magnetostriction values amongst all the compounds studied here. Thus, this compound could have compensation of the magnetocrystalline anisotropy constants of the Sm^{3+} and Nd^{3+} ions. In the present investigation, the $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ compound possesses a large parallel magnetostriction value of $\lambda_\parallel = -1572 \times 10^{-6}$ and it is significantly larger than that of $Tb_{0.27}Dy_{0.73}Fe_2$ compound. The value of $(\lambda_\parallel - \lambda_\perp)$ at 11 kOe of $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ is found to be -2220×10^{-6}, the largest among the samarium substituted anisotropy compensated compounds and, in fact, the largest ever for a pseudobinary RFe_2 compounds. Based on the theoretical values of λ_{111} at 0 K for $SmFe_2$ and $NdFe_2$, the corresponding value for the $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ compound was calculated to be -2576×10^{-6}. The experimental value obtained is in accordance

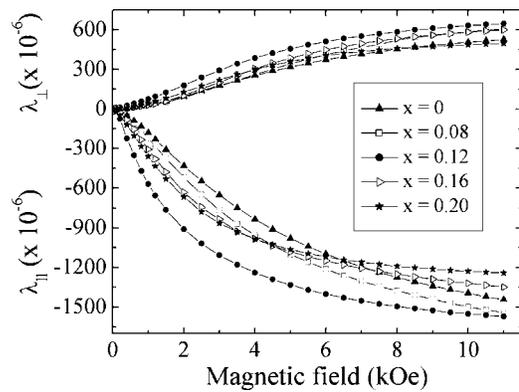with this value. The magnetostriction values at 11 kOe are found to decrease with the higher Nd content. This could be due to the increase of positive magnetostriction values from the Nd sublattice with its higher content.

The XRD patterns of magnetically aligned powders of the $Sm_{1-x}Nd_xFe_{1.93}$ ($x=0$, 0.12, 0.20, 0.24, 0.32, and 0.36) compounds are shown in Fig. 5. The compounds with $0 \leq x \leq 0.20$ reveal intense reflections corresponding to the (111), (222), and (333) planes with the peaks corresponding to all other planes having very small intensities. Thus, for the above compounds, the EMD aligns along the $\langle 111 \rangle$ directions and could be due to the large Sm (having $\langle 111 \rangle$ as the EMD) content. On the other hand, the compounds of $x=0.32$ and above reveal intense peaks corresponding to the (220) planes, indicating that the above compounds could have their EMD corresponding to the $\langle 110 \rangle$ planes. In principle, the Nd rich compounds would have the EMD along $\langle 100 \rangle$. But it is well known that the Nd rich compounds will not be stabilized as a single phase for higher Nd content. Therefore, the EMD of the $Sm_{1-x}Nd_xFe_{1.93}$ compounds may be expected to continuously rotate from $\langle 111 \rangle$ (Sm rich) to $\langle 100 \rangle$ (Nd rich) through the intermediate $\langle 110 \rangle$ and $\langle 11w \rangle$, $\langle 1vw \rangle$, etc. directions, as seen from the XRD pattern of the aligned sample of $Sm_{0.76}Nd_{0.24}Fe_{1.93}$.
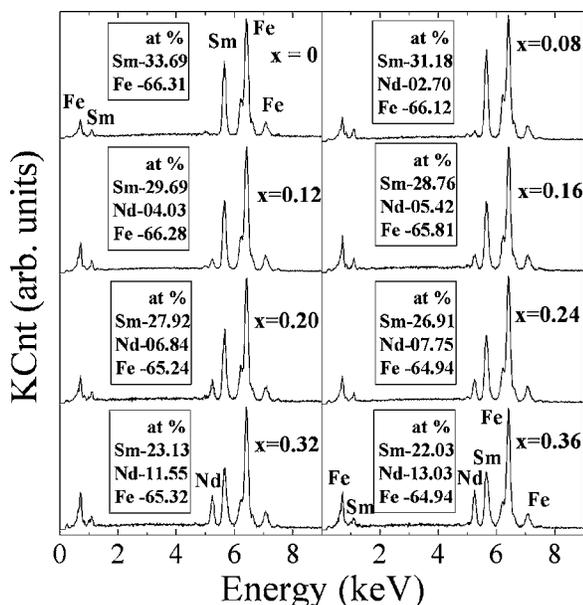

FIG. 3. EDAX patterns of $Sm_{1-x}Nd_xFe_{1.93}$ compounds.
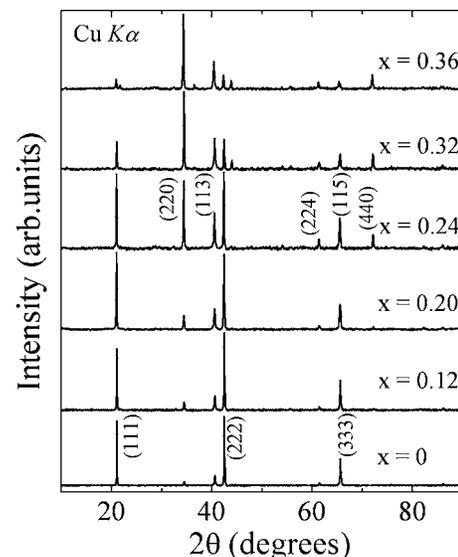

FIG. 5. XRD patterns of magnetically aligned powders of $Sm_{1-x}Nd_xFe_{1.93}$ compounds.
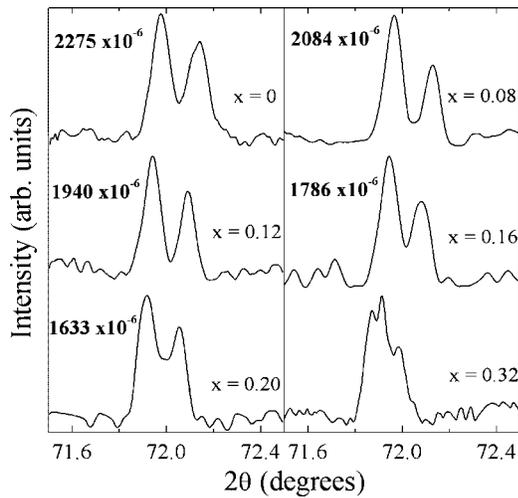
FIG. 6. XRD patterns of (440) reflections of $Sm_{1-x}Nd_xFe_{1.93}$ compounds.

The large spontaneous magnetostriction of some of the $R\mathrm{Fe}_2$ compounds leads to either rhombohedral, orthorhombic, or tetrahedral distortions for the cubic Laves phase compounds whose EMDs are along $\langle 111 \rangle$, $\langle 110 \rangle$, and $\langle 100 \rangle$, respectively.[9,10] Several authors[11,12] have reported that the (440) peaks in rhombohedrally distorted compounds split into $(\bar{4}40)$ and $(4\bar{4}0)$ peaks. If θ, θ_1, and θ_2 are the Bragg angles for the (440), $(\bar{4}40)$, and $(4\bar{4}0)$ reflections, respectively, then the spontaneous magnetostriction is

$$\lambda_{111} = \frac{\Delta\theta}{\tan\theta}, \quad \text{where} \quad \theta = \frac{\theta_1 + \theta_2}{2} \quad \text{and} \quad \Delta\theta = |\theta_1 - \theta_2|.$$

In order to determine the spontaneous magnetostriction induced rhombohedral distortion in the $Sm_{1-x}Nd_xFe_{1.93}$ compounds, XRD patterns of (440) reflections were taken at slow scanning rates (angle 2θ: 71.5°–72.5°; step width: 0.004° and time per step: 60 s) and are shown in Fig. 6. The contribution from the $K\alpha_2$ radiation was removed using 'X' pert HIGH-SCORE PLUS software provided by M/s PANalytical. The splitting of the (440) peaks for the $0 \leq x \leq 0.20$ compounds are seen from Fig. 6. The spontaneous magnetostriction (λ_{111}) values for these compounds were calculated using the above equation and are listed in Fig. 6. The λ_{111} values are found to decrease with increase in the Nd content. The EMD in the above compounds is along the $\langle 111 \rangle$ directions. On the other hand, the (440) peak for the $x=0.32$ compound is seen

to split into three peaks indicating an orthorhombic distortion in this compound supporting the fact that the EMD is along $\langle 110 \rangle$ in this compound. A λ_{111} value of 2275×10^{-6} is observed for the $x=0$ compound. Barbara et al.,[13] reported that $\mathrm{SmFe_2}$ has a negative rhombohedral distortion with λ_{111} value of -2.3×10^{-3}. The $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ compound is found to have a λ_{111} value of 1940×10^{-6}, to the best of our knowledge, the largest ever reported for an anisotropy compensated (at room temperature) pseudobinary cubic Laves phase compound.

In conclusion, the $Sm_{0.88}Nd_{0.12}Fe_{1.93}$ compound is found to exhibit giant magnetostriction values of $\lambda_{\parallel} = -1572 \times 10^{-6}$ and $(\lambda_{\parallel} - \lambda_{\perp}) = -2220 \times 10^{-6}$ due to the anisotropy compensation between Sm^{3+} and Nd^{3+} ions. The EMD of the above compound was determined to be along $\langle 111 \rangle$. The splitting of (440) peak accompanied by the rhombohedral distortion of the above compound was observed through the slow scanned XRD patterns of (440) reflections and λ_{111} is calculated to be 1940×10^{-6}.

[1] A. E. Clark, *Ferromagnetic Materials*, edited by E. P. Wohlforth (North-Holland, Amsterdam, 1980), Vol. 1, p. 531.
[2] N. C. Koon, C. M. Williams, and B. N. Das, J. Magn. Magn. Mater. **100**, 173 (1991).
[3] H. T. Savage, R. Abbundi, A. E. Clark, and O. D. McMasters, J. Magn. Magn. Mater. **15-18**, 609 (1980).
[4] R. Abbundi, A. E. Clark, and O. D. McMasters, J. Appl. Phys. **53**, 2664 (1982).
[5] Z. J. Guo, Z. D. Zhang, B. W. Wang, and X. G. Zhao, Phys. Rev. B **61**, 3519 (2000).
[6] Y. G. Shi, S. L. Tang, Y. J. Huang, L. Y. Lv, and Y. W. Du, Appl. Phys. Lett. **90**, 142515 (2007).
[7] C. Meyer, F. Hartmann-Boutron, J. J. Capponi, J. Chappert, and O. Massenet, J. Magn. Magn. Mater. **15-18**, 1229 (1980).
[8] K. R. Dhilsha, G. Markandeyulu, and K. V. S. Rama Rao, J. Appl. Phys. **70**, 4450 (1991).
[9] A. E. Dwight and C. M. Kimball, Acta Crystallogr., Sect. B: Struct. Crystallogr. Cryst. Chem. **B30**, 2791 (1974).
[10] V. V. Aleksandryan, R. Z. Levitin, A. S. Markosyan, V. V. Snegirev, and A. D. Shchurova, Sov. Phys. JETP **65**, 502 (1987).
[11] W. J. Ren, Z. D. Zhang, X. G. Zhao, W. Liu, and D. Y. Geng, Appl. Phys. Lett. **84**, 562 (2004).
[12] S. N. Jammalamadaka, G. Markandeyulu, and K. Balasubramaniam, J. Appl. Phys. **101**, 09C504 (2007).
[13] B. Barbara, J. P. Giraud, J. Laforest, R. Lemaire, E. Siaud, and J. Schweizer, Physica B & C **86**, 155 (1977).