Study of Spin-Reorientation in Tm$_2$Fe$_{17-x}$Ga$_x$ and Sm$_2$Fe$_{17-x}$Ga$_x$


Abstract—In order to compare their anisotropy behavior, Tm$_2$Fe$_{17-x}$Ga$_x$ and Sm$_2$Fe$_{17-x}$Ga$_x$ ($0 \leq x \leq 8$) powders were magnetically aligned using either static-field or dynamic-field methods. Magnetization measurements were carried out along both the easy and hard magnetic directions for temperatures $10 \leq T \leq 350$ K and fields up to 50 kOe. Spin-reorientation behavior was observed in all of the Tm$_2$Fe$_{17-x}$Ga$_x$ samples studied; the nature of the anisotropy depends on both the Ga content and temperature. In Tm$_2$Fe$_{17-x}$Ga$_x$ for $x = 0, 1, 3, and 5$, the anisotropy changes from easy-uniaxial to easy-plane with increasing temperature; while for $x = 7$ and 8, the anisotropy changes from easy-plane to uniaxial. In contrast, none of the Sm$_2$Fe$_{17-x}$Ga$_x$ samples studied showed spin-reorientation behavior. For Sm$_2$Fe$_{17-x}$Ga$_x$ with $x = 0, 6, 7, and 8$, the anisotropy is easy-plane, while with $x = 1, 2, 3, 4, and 5$, the anisotropy is uniaxial; the nature of the anisotropy does not change with temperature over the range of measurement. The results are discussed in terms of the effect of Ga substitution on the Tm-sublattice and Sm-sublattice anisotropies.

Index Terms—Anisotropy, Sm$_2$Fe$_{17-x}$Ga$_x$, spin-reorientation, Tm$_2$Fe$_{17-x}$Ga$_x$.

I. INTRODUCTION

Although considerable work has been carried out on the R$_2$Fe$_{17-x}$Ga$_x$ compounds and their carbides and nitrides, the physical reasons for the improvements in the magnetic properties remains unclear, particularly with regard to the magnetocrystalline anisotropy. In an early work, the nature of the total magnetocrystalline anisotropy for R$_2$Fe$_{17-x}$Ga$_x$, where R=Y, Sm, Gd, Tb, Dy, Ho, Er, and Tm, was investigated by means of x-ray diffraction on powders which were aligned in a static magnetic field [1]. It was found that the Sm-containing system shows very distinct behavior from the other R-containing systems at room temperature. For R$_2$Fe$_{17-x}$Ga$_x$, with R other than Sm (e.g., R=Tm), x must be greater than 5 in order to change the anisotropy from planar to uniaxial, and above $x = 5$, the anisotropy remains uniaxial up to the largest permitted substitution of $x \approx 8$. However, for Sm$_2$Fe$_{17-x}$Ga$_x$, a relatively small substitution of Ga for Fe ($x \approx 1$) causes the anisotropy to change from planar to uniaxial; a large Ga content ($x \approx 6$) causes the anisotropy to change back to planar again. In a later work, a separation of the total anisotropy for Sm$_2$Fe$_{17-x}$Ga$_x$ ($0 \leq x \leq 8$) into contributions from both the Sm-sublattice anisotropy and Fe-sublattice anisotropy was achieved by magnetization measurements on samples that were aligned using either static-field and dynamic-field methods [2]. The Fe sublattice anisotropy was obtained from corresponding measurements on Y$_2$Fe$_{17-x}$Ga$_x$, ($0 \leq x \leq 8$), as the presence of Y eliminates the rare-earth sublattice contribution.

In addition to separating the anisotropy into its sublattice contributions and determining the variation of these contributions with Ga substitution, magnetization measurements on aligned samples provide the opportunity to study spin reorientation directly. In these systems, a spin reorientation can occur because of the competition between the R-sublattice anisotropy and the negative (easy-plane) Fe-sublattice anisotropy. Since the two sublattice anisotropies normally possess different temperature dependences, the dominant sublattice anisotropy may change enough at a certain temperature, called the spin-reorientation temperature, $T_{spr}$, resulting in a sudden exchange of the easy and hard magnetic directions. In this work, magnetization measurements have been carried out on powder samples of Tm$_2$Fe$_{17-x}$Ga$_x$ and Sm$_2$Fe$_{17-x}$Ga$_x$, for $0 \leq x \leq 8$ which were aligned by either a static-field or dynamic-field method. The results show that the nature of the anisotropy for Sm$_2$Fe$_{17-x}$Ga$_x$ does not change with temperature over the range of measurement from 350 K down to 10 K for $0 \leq x \leq 8$. In contrast, spin-reorientation behavior was observed for all of the Tm$_2$Fe$_{17-x}$Ga$_x$ samples. The results are discussed in terms of the various sublattice contributions.

II. EXPERIMENTAL APPARATUS AND PROCEDURE

Powder samples of Sm$_2$Fe$_{17-x}$Ga$_x$ and Tm$_2$Fe$_{17-x}$Ga$_x$ with $0 \leq x \leq 8$ for both systems were prepared as described previously [1], [3]. X-ray diffraction analysis showed that all of the Sm$_2$Fe$_{17-x}$Ga$_x$ samples are single phase with the rhombohedral (Tb$_2$Zn$_{17}$) structure. Sm$_2$Fe$_{17-x}$Ga$_x$ samples with $x \leq 3$ have the hexagonal (Tb$_2$Ni$_{17}$) structure and samples with $x \geq 5$ have the rhombohedral (Tb$_2$Zn$_{17}$) structure. The Tm$_2$Fe$_{12}$Ga$_4$ sample showed peaks indicating the coexistence of both structures. The powders used in this work with grain sizes less than 20 µm were mixed with epoxy resin and then aligned in a magnetic field at room temperature. Depending on the type of room-temperature anisotropy for the particular sample, either a static-field or dynamic-field procedure was used [4], [5]. For the uniaxial anisotropy samples, a static-field alignment was performed in a dc magnetic field of 15 kOe. For
the planar anisotropy samples, a dynamic-field alignment was used in which the sample was rotated back and forth through an angle of 60° in a dc field of 15 kOe, where the field lies in the plane of the rotation (“stepped oscillation”). Such a procedure makes the basal (easy) planes of different particles parallel (c-axes perpendicular) to the oscillation plane of the sample and, furthermore, the easy axes in the basal plane for these particles are aligned such that they are parallel to one of the two limiting directions of the oscillation. X-ray diffraction confirmed that the alignment procedure was successful. Measurements of the magnetization were made as a function of temperature 10 K ≤ T ≤ 350 K and fields up to 50 kOe using a Quantum Design MPMS SQUID magnetometer.

III. EXPERIMENTAL RESULTS AND ANALYSIS

Fig. 1 shows the magnetization versus temperature behavior for Tm₂Fe₁₇ (x = 0) in a field of 300 Oe. The sample was first zero-field-cooled (ZFC) from room temperature to 10 K, and then the 300 Oe field was applied with its direction lying in the same plane as that used for the sample rotation during the dynamic-field alignment (see text). Measurements were then made as the temperature increased (closed circles) and then decreased (open circles). Spin-reorientation is seen to occur at 84 K; the Curie temperature is near room temperature. For all three curves, the open circles represent the magnetization with the field lying in the rotation plane as described above for Fig. 1, while the closed circles represent the magnetization perpendicular to the plane. It is clear from the curves, that the easy direction is along the c-axes for 10 K and lies in the basal plane for 170 K and 300 K. Again, similar magnetization versus field curves were obtained for the Tm₂Fe₃Ga₅ samples which also have uniaxial anisotropy at room temperature. The results are summarized in Fig. 4 which shows the measured values of Tₘ as a function of the Ga content, x (open circles).
Fig. 3. Magnetic moment versus temperature for Tm$_2$Fe$_{17-x}$Ga$_x$ in a magnetic field of 300 Oe. The sample was first zero-field-cooled from room temperature to 10 K, and then the 300 Oe field was applied with its direction parallel to the static-field alignment direction (see text). Measurements were then made as the temperature increased (closed circles) and then decreased (open circles). Spin-reorientation is seen to occur at 198 K.

Fig. 4. Spin-reorientation temperature $T_{rx}$ versus Ga content $x$ for Tm$_2$Fe$_{17-x}$Ga$_x$ (open circles). Values obtained from ac susceptibility measurements in an earlier work (crosses) [6].

Also shown in Fig. 4 for comparison are the values obtained from ac susceptibility measurements in an earlier work (crosses) [6].

As reported previously, Sm$_2$Fe$_{17-x}$Ga$_x$ with $x = 1, 2, 3, 4$, or 5 is uniaxial and the static-field alignment was performed in a field of 15 kOe at room temperature. For Sm$_2$Fe$_{17-x}$Ga$_x$ with $x = 0, 6, 7$, or 8, which has planar anisotropy, the dynamic-field alignment in 15 kOe as described above was performed. In contrast to the behavior observed for Tm$_2$Fe$_{17-x}$Ga$_x$, no spin-reorientation transition was observed for any of the Sm$_2$Fe$_{17-x}$Ga$_x$ samples over the range of measurement from 10 K to 350 K.

IV. DISCUSSION AND CONCLUSIONS

In summary, this work presents a comparison of the anisotropy behavior for the substitutional systems Tm$_2$Fe$_{17-x}$Ga$_x$ and Sm$_2$Fe$_{17-x}$Ga$_x$ ($0 \leq x \leq 8$). Powder samples were magnetically aligned at room temperature by using either static-field or dynamic-field methods, depending on the nature of the anisotropy (uniaxial or easy-planar, respectively). Magnetization measurements, which were carried out along both the easy and hard magnetic directions, directly demonstrated that spin-reorientation occurs for all of the Tm$_2$Fe$_{17-x}$Ga$_x$ samples studied; the nature of the anisotropy depends on both the Ga content and temperature. In Tm$_2$Fe$_{17-x}$Ga$_x$ for $x = 0, 1, 3$, or 5, the anisotropy changes from uniaxial to easy-plane with increasing temperature; while for $x = 7$ or 8, the anisotropy changes from easy-plane to uniaxial. On the other hand, the results show that Sm$_2$Fe$_{17-x}$Ga$_x$ samples with $x = 0, 6, 7$, and 8 possess easy-plane anisotropy while samples with $x = 1, 2, 3, 4$, and 5 possess uniaxial anisotropy, and the nature of the anisotropy does not change with temperature for 10 K $\leq T \leq 350$ K.

These experiments show that Ga has a much stronger effect on the Sm-sublattice anisotropy than on the Tm-sublattice anisotropy. In a previous work referred to above, the total anisotropy for Sm$_2$Fe$_{17-x}$Ga$_x$ ($0 \leq x \leq 8$) was separated into contributions from both the Sm-sublattice and Fe-sublattice by magnetization measurements on aligned samples [2]. It can be seen from Fig. 3 in [2] that the Sm-sublattice anisotropy dominates the Fe-sublattice anisotropy for $x \geq 1$. It increases abruptly with Ga content, reaching a (broad) maximum of $1.8 \times 10^7$ erg/cm$^3$ for $x \approx 2$ to 3, and then decreases and becomes negative for $x \approx 5.5$. Consequently, because of this domination, no spin-reorientation behavior occurs in Sm$_2$Fe$_{17-x}$Ga$_x$ for the compositions studied, and over the temperature range of measurement. For the Tm$_2$Fe$_{17-x}$Ga$_x$ system, however, the competition between the nearly equal Tm-sublattice and Fe-sublattice anisotropy results in the occurrence of spin-reorientation behavior. Based on this study, it is anticipated that a spin-reorientation will take place in Sm$_2$Fe$_{17-x}$Ga$_x$ within very narrow ranges of Ga content for $x \leq 1$ and $x$ between 5 and 6.

REFERENCES