Magnetic properties of Sm2(Fe1-xGax)17(x=0-0.5) compounds and their nitrides

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Magnetic properties of \( \text{Sm}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) \((x=0-0.5)\) compounds and their nitrides

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Magnetic properties of \( \text{Sm}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) compounds and their nitrides have been studied. Substitution of Ga for Fe leads to an increase in lattice constants. Introduction of nitrogen results in a further increase in lattice constants. Substitution of Ga for Fe causes a dramatic change of the Curie temperature of the \( \text{Sm}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) compounds. When \( x=0.2 \) the Curie temperature is enhanced by about 200 K. X-ray-diffraction patterns of aligned samples of \( \text{Sm}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) compounds show that alloys with \( x=0.15, 0.20, \) and 0.25 exhibit uniaxial anisotropy at room temperature. The introduction of nitrogen made the samples with \( x=0.4 \) exhibit uniaxial anisotropy at room temperature. The Curie temperature of the nitrides decreases with the Ga concentration. The anisotropy fields of the nitrides derived from the high-field magnetization. The changes of the magnetic anisotropy, saturation magnetization, and the moment of the Fe atoms in the nitrides and their parent compounds with Ga concentration are discussed.

I. INTRODUCTION

Recently many studies on improving the magnetic properties of \( \text{R}_2\text{Fe}_{17} \) compounds have been performed. The most striking improvements, the strong enhancements of the Curie temperature and uniaxial anisotropy, have been achieved by absorption of nitrogen.\(^1\) The magnetic ordering temperature and other magnetic properties can also be improved by uptaking hydrogen or carbon as well as by substituting some elements such as Al, Si, and Co for Fe in \( \text{R}_2\text{Fe}_{17} \) compounds.\(^2\)\(^-\)\(^7\)

In the present work we have focused our attention on the crystal structure and magnetic properties of \( \text{Sm}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) \((x=0-0.5)\) compounds and their nitrides, especially on magnetization and magnetocrystalline anisotropy. The effects of Ga substitution for Fe and of the introduction of interstitial nitrogen on the Curie temperature, the magnetic anisotropy, and the saturation magnetization have been determined.

II. EXPERIMENTAL METHODS

All \( \text{Sm}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) host compounds with \( x=0, 0.01, 0.02, 0.04, 0.07, 0.1, 0.15, 0.2, 0.25, 0.3, 0.35, 0.4, \) and 0.5 were prepared by arc melting. The nitrides were formed by heating the powder samples of \( \text{Sm}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) compounds in a mixture of \( \text{NH}_3 \) gas and \( \text{H}_2 \) gas under a pressure of 1 atm at 720 K for 20 min.

X-ray diffraction was employed to determine the structure, phase composition, the lattice parameters, and the anisotropy of aligned samples. The thermomagnetic \( \sigma-T \) curves were measured by means of a vibrating sample magnetometer in a field of 0.05 T. The Curie temperatures \( T_C \) were derived from \( \sigma-T \) plots.

The high-field magnetization curves were measured at 4.2 K in high fields up to 21 and 35 T for the host compounds and their nitrides, respectively, at the University of Amsterdam.\(^8\) The anisotropy fields of the nitrides were derived from the intersection point of the high-field magnetization curves measured with the field applied parallel and perpendicular to the aligned direction. The saturation magnetization was deduced from \( \sigma_x-1/B \) plots.

III. RESULTS AND DISCUSSION

Based on x-ray powder diffraction, all the investigated \( \text{Sm}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) compounds and their nitrides crystallize in the \( \text{Th}_2\text{Zn}_{17} \)-type structure. A small amount of impurity was found in a few samples. Ga substitution for Fe does not change the structure of \( \text{Sm}_2\text{Fe}_{17} \), but leads to an expansion of the unit cell (Fig. 1). This result may be ascribed to the larger atomic radius of Ga atom compared to Fe. After introduction of nitrogen, the volume of the unit cell further increases; but, the magnitude of increase is smaller than that resulting from the Ga substitution. The nitrogen content introduced into the compounds, determined by weighing, decreases linearly with Ga concentration from 2.6 for \( x=0 \) to 1 for \( x=0.5 \). One can conclude that the substitution of Ga for Fe prevents the introduction of nitrogen.

The Curie temperatures of \( \text{Sm}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) compounds and their nitrides are shown in Fig. 2. This figure clearly displays that the Curie temperature of \( \text{Sm}_2(\text{Fe}_{1-x}\text{Ga}_x)_{17} \) compounds first goes up, passes through a maximum at about \( x=0.2 \), then decreases with increasing Ga content. The initial increase of \( T_C \) is mainly due to the volume expansion. At higher Ga concentration, the average iron magnetic moment decreases dramatically and causes a decrease in \( T_C \).\(^9\) This variation of \( T_C \) may also be associated with preferential substitution of Ga atoms for Fe atoms.\(^9\) Ga atoms preferentially substitute for Fe atoms at the sites responsible for negative exchange interaction. The Curie temperature of the nitrides decreases monotonically with Ga concentration.
When $x \leq 0.2$, the Curie temperature of the nitrides is higher than that of their parent compounds, this indicates that the introduction of N atoms results in an enhancement of the exchange interaction.\(^{10}\)

X-ray-diffraction patterns at room temperature with CoK\(\alpha\) radiation for the aligned samples of \(\text{Sm}_x(\text{Fe}_{1-x}\text{Ga})_{17}\) show that the samples with $x=0.15, 0.20,$ and 0.25 exhibit uniaxial anisotropy; the others are planar [Fig. 3(a)]. X-ray-diffraction patterns of \(\text{Sm}_x(\text{Fe}_{1-x}\text{Ga})_{17}N_y\) [Fig. 3(b)] show that the samples with $x \leq 0.3$ exhibit uniaxial anisotropy at room temperature. Ga substitution for Fe in \(\text{Sm}_2(\text{Fe}_{1-x}\text{Ga})_{17}\) compounds enhances the transition temperature of the anisotropy from easy plane to easy c axis.

This may be attributed to a change of the second crystal-field coefficients $A_{20}$ at the rare-earth sites in \(\text{Sm}_x(\text{Fe}_{1-x}\text{Ga})_{17}\) compounds toward more negative values. After the uptake of the N atoms, the $A_{20}$ becomes more negative,\(^{11,12}\) which makes the samples of \(\text{Sm}_x(\text{Fe}_{1-x}\text{Ga})_{17}N_y\) with $x \leq 0.3$ have uniaxial anisotropy at room temperature.

The saturation magnetization of \(\text{Sm}_x(\text{Fe}_{1-x}\text{Ga})_{17}\) and their nitrides decreases monotonically with Ga concentration. In order to get more information about the influence of Ga substitution on the magnetization, the average iron magnetic moment $\mu_{Fe}$ was calculated on the basis of the saturation magnetization measured at 4.2 K. In the process of evaluating $\mu_{Fe}$, it was assumed that the magnetic moment of the Sm ion is independent of the Ga concentration, and that its magnetic moment was same as that of free Sm ion. Our results imply that both in the nitrides and in the parent compounds the iron moment $\mu_{Fe}$ decreases monotonically with Ga concentration. The average iron moment decreases with Ga concentration from $2.11 \mu_B$ for $x=0$ to $1.51 \mu_B$ for $x=0.5$ for the nitrides and from $1.96 \mu_B$ for $x=0$ to $1.54 \mu_B$ for $x=0.5$ for the parent compounds. This is a reduction of the Fe moment.
due to the substitution of Ga, which is very similar to the case of Al substitution for Fe in Sm$_2$(Fe$_{1-x}$Al$_x$)$_{17}$.

Introduction of nitrogen leads to a change of the magnetic anisotropy of Sm$_2$Fe$_{17}$ from planar to uniaxial in the whole temperature range up to $T_c$. Substitution of Ga for Fe also significantly influences the anisotropy of the nitrides. Figure 4 shows the anisotropy field $B_a$ as a function of Ga concentration for the Sm$_2$(Fe$_{1-x}$Ga$_x$)$_{17}$N$_y$ compounds. For $x \leq 0.3$, $B_a$ decreases very slowly with Ga concentration. Obviously there is a contribution to the uniaxial anisotropy resulting from the substitution of Ga for Fe, for $x > 0.3$, $B_a$ decreases quickly. $B_a$ becomes zero at $x = 0.4$, which is in good agreement with the results of the x-ray diffraction mentioned above. It is well known$^{13}$ that in Sm$_2$Fe$_{17}$N$_y$ compounds the N atoms preferentially occupy 9e sites which are the nearest to the Sm atoms. The N atoms influence the crystal field at the Sm sites resulting in a significant increase of $A_{20}$ of the Sm atoms,$^{11,12}$ so that the uniaxial anisotropy of the Sm sublattice increases. Ga substitution for Fe prevents the introduction of N atoms, so that the anisotropy field of Sm$_2$(Fe$_{1-x}$Ga$_x$)$_{17}$N$_y$ decreases with Ga concentration.

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