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Annealing effects on the magnetic properties of $Nd_2Fe_{17-x}Ga_x$

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Abstract

Magnetization of $Nd_2Fe_{17-x}Ga_x$, where $0 \leqslant x \leqslant 3$, has been measured at temperatures between 5 and 300 K and in fields between 0 and 50 kOe. These samples have the rhombohedral Th_2Zn_{17} structure. Magnetization studies at 5 K indicate that the saturation magnetization decreases with increasing Ga concentration while at 300 K it increases for $x > 0$ reaching a maximum at $x = 1$, and decreases for $x > 1$. Samples annealed for different times are found to have different magnetization for the same value of x. The low-field magnetization measurements show irreversible behavior which is a reflection of hysteretic effects in this system. The peak in the magnetization shifts to higher temperatures upon increasing the Ga amount due to the increase in the Curie temperature. © 2000 Published by Elsevier Science B.V. All rights reserved.

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1. Introduction

Enhancement of the magnetic properties of the R_2Fe_{17} compounds can be achieved by substitution of Ga, Al, Si, V, Co, and Ni for Fe [1–10] and/or by interstitial addition of nitrogen, carbon or hydrogen [11–15]. Substituting Ga for Fe in the $R_2Fe_{17-x}Ga_x$ compounds was found to expand the unit cell and thus increase the Curie temperature T_c, which is due to the magnetovolume effect and changes in the hybridization. Shen et al. [5] studied the $Sm_2Fe_{17-x}Ga_x$ system and found that T_c increases with x and reaches a maximum of 595 K for $x = 3$; they also observed that the magnetic anisotropy changes from basal-plane for $x = 0$ to uniaxial for $x \geqslant 2$. Long et al. [16] found that Curie temperature for $Ce_2Fe_{17-x}Ga_x$ increases from 240 K for $x = 0$ to 430 K for $x = 2$. We studied the structural properties of $Nd_2Fe_{17-x}Ga_x$ [17] and found that these samples crystallize in the rhombohedral Th_2Zn_{17} structure and they have in-plane anisotropy for all values of x and for all the investigated annealing times. First-principle studies by Sabiryanov and Jaswal [18,19] give T_c in very good agreement with the experimental data for 2–17 compounds and their variants. They find that Fe–Fe exchange interactions depend on the environment in a complicated manner.

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In this paper we study the effect of Ga substitution and annealing time on the magnetic properties of $Nd_2Fe_{17-x}Ga_x$ compounds.

2. Experimental procedure

Bulk samples of $Nd_2Fe_{17-x}Ga_x$ with $x = 0, 1, 2,$ and 3 were prepared by arc-melting the elements of at least 99.9% purity in a water-cooled copper boat in a flowing-argon gas atmosphere. The alloys were melted four-to-five times to insure homogeneity. After melting the samples they were cut into three pieces, wrapped separately in tantalum foil, heat-treated in vacuum below 3×10^{-6} Torr pressure at 925°C for 3 days (A), 22 days (B), or 42 days (C), and subsequently quenched in water. The phase purity for all the samples was determined by X-ray diffraction using Cu K_α radiation and showed only the rhombohedral Th_2Zn_{17} structure for samples with $x = 0, 1, 2,$ and 3 annealed for different times. The magnetization of the alloys was measured by a superconducting quantum interference device (SQUID) magnetometer in the temperature range 5–300 K and in fields from 0 to 50 kOe.

3. Results and discussion

Fig. 1 shows a typical initial magnetization curve ($M(H)$) for $Nd_2Fe_{15}Ga_2$ annealed for 42 days and measured at a temperature of 5 K. From this figure we see that the sample is magnetically ordered. We find the saturation magnetization (M_s) at $T = 5$ and 300 K by using the law of approach to saturation, i.e., by plotting M versus $1/H$ and extrapolating M to $(1/H) = 0$. The saturation magnetization for the samples annealed for 3 days (A), 22 days (B), and 42 days (C) and measured at 5 K is shown in Fig. 2. This figure shows that the saturation magnetization decreases with increasing Ga concentration (X) for all samples annealed for different times. This is in qualitative agreement with magnetization values, magnetic moments, and hyperfine fields for other similar compounds [16]. The difference between the magnetization values for samples with the same value of x but different annealing times might be attributed to variations in the degree of ordering which can affect site-dependent moment values through local environment effects.

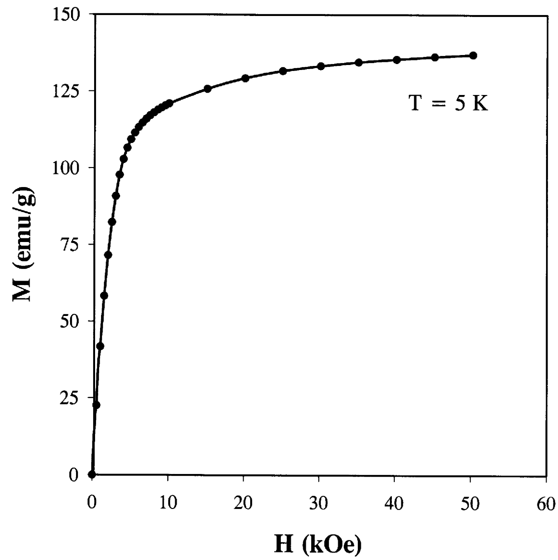

Fig. 1. Typical initial magnetization curve for $Nd_2Fe_{15}Ga_2$ annealed for 42 days and measured at a temperature of 5 K.

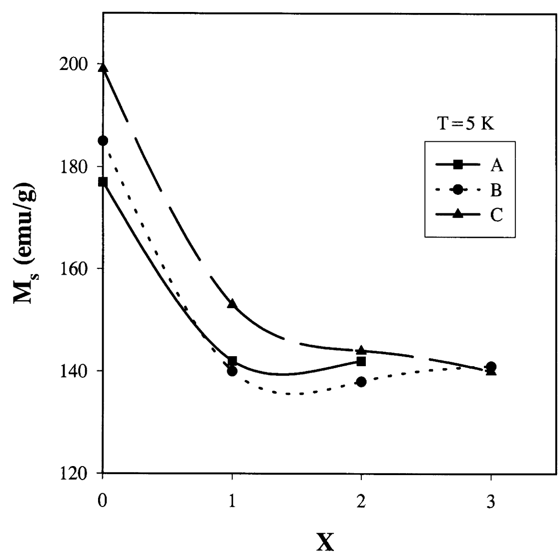

Fig. 2. Saturation magnetization for $Nd_2Fe_{17-x}Ga_x$ compounds annealed for 3 days (A), 22 days (B), and 42 days (C) and measured at 5 K.

Fig. 3 shows the saturation magnetization for the A and B series of samples at a temperature of

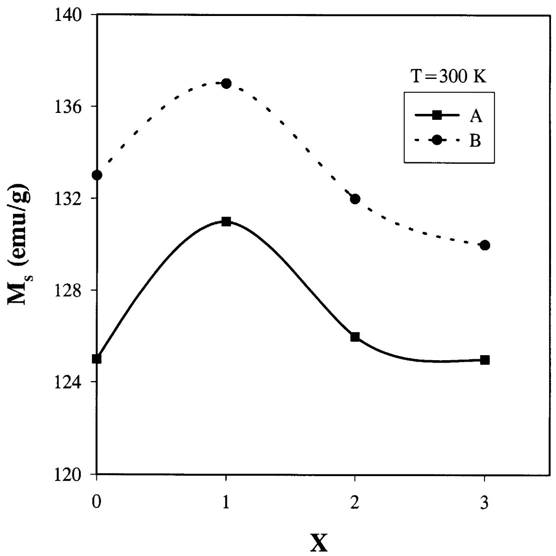

Fig. 3. Saturation magnetization at 300 K for $Nd_2Fe_{17-x}Ga_x$ compounds annealed for 3 days (A), and 22 days (B).

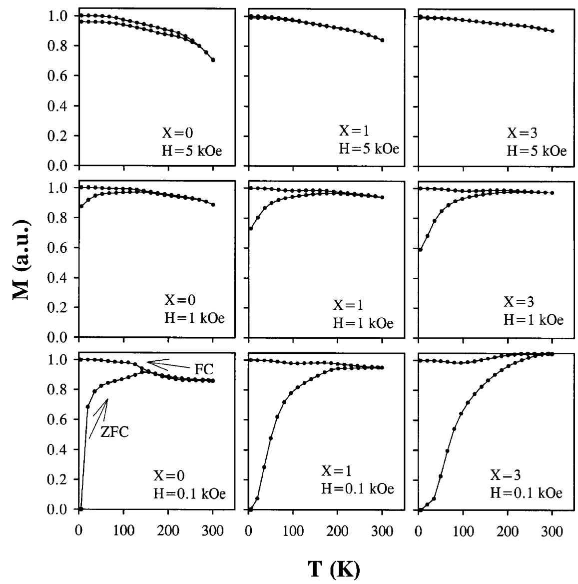

Fig. 4. Temperature dependence of zero-field cooled (ZFC) and field cooled (FC) magnetization (M) for $Nd_2Fe_{17-x}Ga_x$ compounds at applied fields of 0.1, 1, and 5 kOe.

300 K. From this figure we see that the saturation magnetization for samples annealed for 22 days is higher than the saturation magnetization for samples annealed for 3 days. This difference can be attributed to the difference in the local environments as discussed above. The increase in the saturation magnetization for small values of x is due to the increase in T_c caused by the expansion of the unit cell by Ga substitutions in these compounds which lowers the average Fe–Fe hybridization and hence increases the Curie temperature. The Curie temperature for the parent compound is about 380 K and it increases to 600 K for the compound with $x = 3$ [20]. The saturation magnetization for $x > 1$ decreases due to the increasing concentration of the non-magnetic element (Ga). This behavior is in qualitative agreement with observations for other similar compounds [15].

The results of constant-field magnetic measurements for samples annealed for 42 days and for $x = 0$, 1, and 3 are shown in Fig. 4, where the magnetic measurements on samples demagnetized at room temperature and cooled to 5 K in zero field [zero-field cooled (ZFC)] were made in a field of 0.1, 1, or 5 kOe as the temperature was raised to 300 K. The temperature was then reduced to 5 K and the magnetization (M) was measured with the same fields [field cooled (FC)]. The results for all values of x studied show the existence of irreversible phenomena as seen from the separation of the FC and ZFC curves at $H = 0.1$, and 1 kOe. This behavior essentially disappears at $h = 5$ kOe (for comparison purposes, the coercivities at $T = 5$ K are 0.2, 0.4, and 0.6 kOe for $x = 0$, 1, and 3, respectively). The observed irreversible effects at low fields can be attributed to the normal hysteretic effects (Hopkinson effect) in magnetic materials. The shift in the magnetization peak in Fig. 4 to a higher temperature with increasing Ga concentration is due to the increase in the Curie temperature.

4. Conclusion

Magnetic properties of $Nd_2Fe_{17-x}Ga_x$ have been studied by magnetization measurements. Measurements at room temperature show that samples with $x = 0, 1, 2$, and 3 are ferromagnetic. We find that the room-temperature (RT) saturation magnetization for these samples increases with increasing Ga concentration reaching a maximum at $x = 1$, and then decreases. We also find that the RT saturation magnetizations of samples annealed for longer times are higher than those of samples annealed for shorter times, which may be due to changes in the degree of order. Low-temperature (5 K) measurements show that the saturation magnetization for all samples studied decreases with increasing x. This is similar to what has been observed for other systems. Zero-field cooled and field cooled magnetization measurements show irreversible behavior due to hysteretic effects. Understanding and controlling the increase in the saturation magnetization and Curie temperature with substitution in these or related compounds is important for the development of enhanced permanent-magnet materials.

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