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## Influence of Ga on the Fe anisotropy in $Y_2Fe_{17-x}Ga_x$

Ll M Martínez†, J S Muñoz†, V Skumryev†‡, Nora Sheludko‡,  
 Bao-gen Shen§, Xiao-Lei Rao|| and J M D Coey||

† Department of Physics, Universitat Autònoma de Barcelona, Barcelona, Spain

‡ Faculty of Physics, University of Sofia, 1126 Sofia, Bulgaria

§ Institute of Physics, Chinese Academy of Sciences, PO Box 603, Beijing, People's Republic of China

|| Department of Pure and Applied Physics, Trinity College, Dublin 2, Ireland

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**Abstract.** First- and second-order anisotropy constants,  $K_1$  and  $K_2$ , have been determined on aligned samples of  $Y_2Fe_{17-x}Ga_x$  ( $x = 0$  to 7.5) compounds by measuring magnetization and torque curves at temperatures between 4.2 and 300 K. Low-field ac susceptibility has been studied from 80 K to room temperatures. It has been found that a partial substitution of Ga for Fe in  $Y_2Fe_{17}$  initially decreases the absolute value of anisotropy constants thus indicating a weakening of the easy plane character of the Fe anisotropy. The Fe anisotropy becomes uniaxial for a Ga concentration slightly above 6.5 atoms  $fu^{-1}$ . A strengthening of the uniaxial anisotropy with further increase of Ga concentration is indicated by an increase in  $K_1$ , determined by torque measurements at room temperatures. The results show the important changes which Fe anisotropy undergoes when Fe atoms are partially substituted by Ga in  $R_2Fe_{17}$  compounds. They can be used to separate the anisotropy contributions from the Fe sublattice and the R sublattice in  $R_2Fe_{17-x}Ga_x$  compounds with magnetic R and thus determine the influence of Ga on the magnetic anisotropy in these materials.

### 1. Introduction

The rare earth–iron compounds of the composition  $R_2Fe_{17}$  have attracted much attention as possible high-performance permanent magnets. In recent years many efforts towards overcoming the two severe drawbacks to the possible application of these materials—their low Curie temperatures and their room-temperature easy plane anisotropies—have been made. The future of  $R_2Fe_{17}$  materials seems bright, as indicated by the discovery of the interstitial compound  $Sm_2Fe_{17}N_x$  [1], which exhibits strong uniaxial magnetic anisotropy and has a Curie temperature 280 K higher than that of the parent compound. More recently it has been found that the substitution of some Ga for Fe in  $R_2Fe_{17}$  can effectively increase the Curie temperature and induce a room-temperature uniaxial anisotropy [2–10].

The anisotropy energy for hexagonal crystals is represented by the phenomenological expression:

$$E_a = K_1 \sin^2 \theta + K_2 \sin^4 \theta + K_3 \sin^6 \theta + K'_3 \sin^6 \theta \cos 6\phi + \dots \quad (1)$$

where  $\theta$  and  $\phi$  are the polar and azimuthal angles of the magnetization relative to the  $c$ -axis of the crystal and  $K_i$  are the anisotropy constants. The terms with  $\sin^6 \theta$  may be neglected. Then according to the magnitudes and signs of  $K_1$  and  $K_2$ , three phases are found: easy axis when  $K_1 > 0$  and  $K_1 + K_2 > 0$ ; easy 'cone' when  $K_1 < 0$  and  $K_1 + 2K_2 > 0$  and easy

plane when  $K_1 > 0$  and  $K_1 + K_2 < 0$ , or  $K_1 < 0$  and  $K_1 + 2K_2 < 0$ . When  $K_1$  dominates the anisotropy energy, then its sign will determine the type of anisotropy:  $K_1$  is negative for easy plane anisotropy and positive for easy axis.

To a first approximation, the total first-order magnetocrystalline anisotropy constant  $K_1$  is a sum of  $K_1(\text{R})$  and  $K_1(\text{Fe})$ , the first anisotropy constant of the rare earth and the iron sublattices, which have a different temperature dependence. It is well established that  $K_1(\text{Fe})$  is negative for  $\text{R}_2\text{Fe}_{17}$  compounds for all temperatures below  $T_c$ . Usually, the R ion anisotropy is expressed by the product of the electric field gradient, represented by the second-order crystal field parameter  $A_2^0$ , and the temperature-dependent quadrupole moment of the rare earth ion which is the product of the second-order Steven coefficient  $\alpha_j$  and the quantity  $3\langle J_z^2 \rangle - J(J+1)$  where  $\langle \rangle$  denotes a thermal average. A negative product of both quantities gives characteristic uniaxial anisotropy with  $K_1(\text{R}) > 0$ .

To explain the change of anisotropy type upon Ga substitution in  $\text{Sm}_2\text{Fe}_{17}$  ( $\alpha_j > 0$  for Sm) an enhancement in the magnitude of  $A_2^0$  is claimed in [2], while in [10] a reversal in sign of  $A_2^0$  is assumed to explain this change in  $\text{R}_2\text{Fe}_{17}$  compounds with R having negative  $\alpha_j$ . However, x-ray study [6] indicates that a change of anisotropy type takes place also for  $\text{Y}_2\text{Fe}_{17-x}\text{Ga}_x$  (Y is non-magnetic)—a fact which emphasizes that the change in the Fe sublattice anisotropy upon Ga substitution must be also taken into account in analysing the anisotropy in  $\text{R}_2\text{Fe}_{17-x}\text{Ga}_x$  compounds. Here we report the anisotropy constants of the Fe sublattice in the Y compounds as a function of Ga concentration and temperature.

## 2. Experiment

$\text{Y}_2\text{Fe}_{17-x}\text{Ga}_x$  ( $x = 0, 1, 2, 3, 4, 5, 6, 6.5, 7, 7.5$ ) ingots were prepared by arc melting and subsequent annealing as described in [6]. The ingots were homogenized at 1273 K in an argon atmosphere for 24 h and then quenched to room temperature, resulting in a single-phase compound of the 2:17-type structure. Room-temperature magnetic anisotropy type was determined from x-ray diffraction patterns of aligned samples. For magnetic anisotropy studies cylindrical samples ( $l = 10$  mm,  $d = 3$  mm) were prepared by mixing 15–20% fine powder (particle size under 15  $\mu\text{m}$ ) with epoxy resin, followed by alignment in a 1 T magnetic field in two different ways, depending on the type of room-temperature magnetic anisotropy. For alignment of easy axis materials ( $x = 7$  and 7.5) and for easy cone material ( $x = 6.5$ ) a static magnetic field was applied. For the easy plane materials ( $x = 0$  to 6) a rotating field was used [11] in a device where a cylindrical tube with the mix is fixed on a rotating axis, connected to a motor, in a static field of an electromagnet applied normal to the axis of rotation. This procedure insures that the magnetically hard  $c$ -axes of all crystallites are collinear. Magnetization curves were measured at fixed temperatures using a SQUID magnetometer equipped with a 5 T superconducting magnet, with the field applied in the easy or hard magnetization direction. Assuming non-interacting particles with a spherical shape the data are corrected using a demagnetization factor  $N = 1/3$ . Torque curves were measured at room temperatures using a torque magnetometer with an electromagnet in a field up to 1.3 T applied in the plane containing the  $c$ -axis. The samples were cut in the form of a disc with the  $c$ -axis along the diameter. While the field is turned around the disc axis the torque is measured by optical means. AC susceptibility as a function of temperature was measured after cooling the samples from room temperatures to 80 K using a Lake Shore susceptometer. The ac field of 80  $\text{A m}^{-1}$  rms and frequency 111 Hz was applied along the cylinder axis to reduce the demagnetizing field effect.

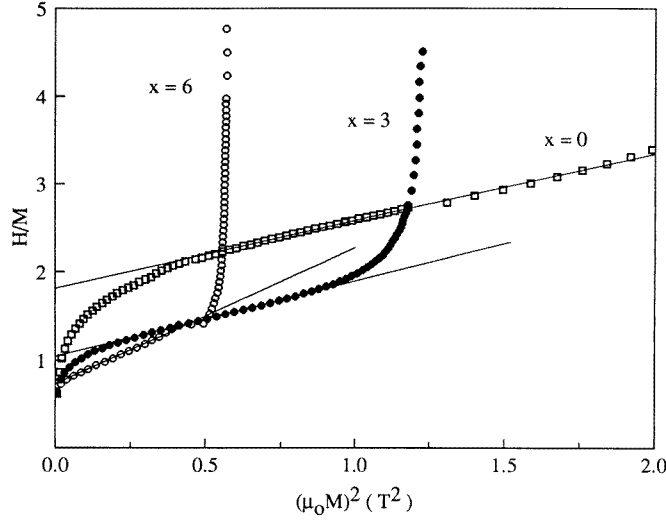


Figure 1. Sucksmith–Thompson plots of  $Y_2Fe_{17-x}Ga_x$  at 4.2 K.

### 3. Results and discussion

#### 3.1. Magnetization curves

The constants  $K_1$  and  $K_2$  are determined from the magnetization curves, measured in the hard direction, using the Sucksmith–Thompson method [12]. When the magnetization processes are determined only by domain magnetization rotation, which is the dominant mechanism when the field is applied in the hard magnetization direction, and if only  $K_1$  and  $K_2$  are considered the following expressions are valid:

$$\frac{H}{M_c} = -\frac{2K_1 + 4K_2}{\mu_0 M_s^2} + \frac{4K_2}{M_s^4} M_c^2 + N \quad (K_1 < 0) \quad (2)$$

for the case of easy plane anisotropy, and

$$\frac{H}{M_{ab}} = \frac{2K_1}{\mu_0 M_s^2} + \frac{4K_2}{\mu_0 M_s^4} M_{ab}^2 + N \quad (K_1 > 0) \quad (3)$$

for the case of easy axis anisotropy, where  $M_c$  and  $M_{ab}$  are the magnetization along the crystallographic  $c$ -axis and the  $ab$ -plane,  $H$  is the applied field,  $N$  the demagnetizing factor and  $M_s$  is the saturation magnetization.

$M_s$  can be easily determined from the magnetization curves using the law of approach to saturation. Plotting  $M^2$  against  $H/M$  using the first-quadrant data, a linear region is observed, as shown in figure 1 for some selected curves. The existence of a linear part in Sucksmith–Thomson plots supports our conjecture of neglecting the higher-order terms in (1). The linear part was fitted by the expression  $H/M = AM^2 + B$ .

To obtain  $K_1$  from the fit, two different expressions should be used. For easy axis samples

$$K_1 = \frac{\mu_0 M_s^2}{2} (B - N) \quad K_2 = \frac{A \mu_0 M_s^4}{4}. \quad (4)$$

For easy plane samples, the expression becomes

$$K_1 = \frac{\mu_0 M_s^2}{2} (N - B - A M_s^2) \quad K_2 = \frac{A \mu_0 M_s^4}{4}. \quad (5)$$

According to these expressions,  $K_2$  is independent of the demagnetizing factor, while  $K_1$  depends on it.

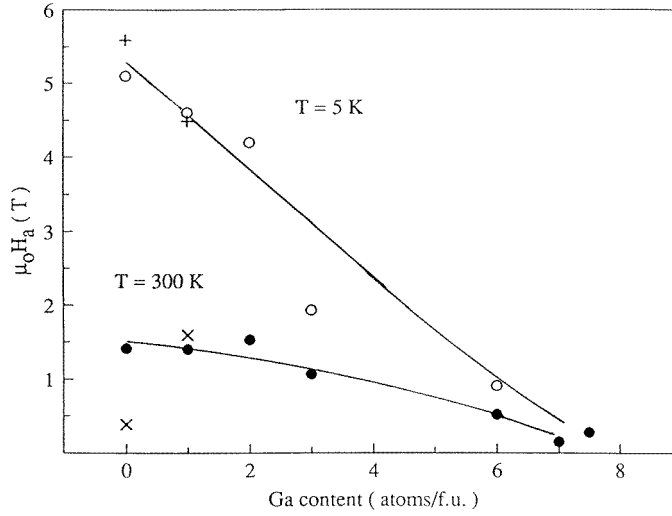
The data obtained for  $K_1$  and  $K_2$  for  $x \leq 6.5$  are listed in table 1. The  $K_1$  data agree with results reported by Li *et al* for  $x < 6$  [13]. As mentioned above they are calculated assuming a demagnetizing factor  $N = 0.33$ , i.e. assuming non-interacting particles with a spherical shape (to take into account the influence of non-spherical shape an error of  $\pm 0.03$  can be included). This should be a good approximation when the concentration of magnetic particles is low. In the opposite case, when the particle concentration is high enough and the interactions between the particles cannot be neglected a demagnetizing factor determined by the sample geometry should be used. In the latter case the calculated  $|K_1|$  values will be about 15% bigger than in the former. An approach for determining  $N$  which takes into account the powder to epoxy ratio has been proposed in [14]. Using this approach values of  $N$  between 0.320 and 0.329 have been found for the different samples, which is very close to the value for non-interacting spherical particles.

**Table 1.** Saturation magnetization and anisotropy constants of  $Y_2Fe_{17-x}Ga_x$ , determined from the magnetization curves.

| Ga content<br>(atoms fu <sup>-1</sup> ) | $T$<br>(K) | $\mu_0 M_s$<br>(T) | $K_1$<br>(MJ m <sup>-3</sup> ) | $K_2$<br>(MJ m <sup>-3</sup> ) |
|---|------------|--------------------|--------------------------------|--------------------------------|
| 0                                       | 5          | 1.71               | -3.47                          | 1.03                           |
|   | 77         | 1.64               | -2.52                          | 0.67                           |
|   | 300        | 1.09               | -0.62                          | 0.16                           |
| 1                                       | 5          | 1.52               | -2.80                          | 0.73                           |
|   | 77         | 1.45               | -2.28                          | 0.73                           |
|   | 300        | 1.10               | -0.62                          | 0.153                          |
| 2                                       | 5          | 1.37               | -2.30                          | 0.65                           |
|   | 77         | 1.35               | -1.72                          | 0.49                           |
|   | 300        | 1.09               | -0.67                          | 0.164                          |
| 3                                       | 5          | 1.23               | -0.95                          | 0.296                          |
|   | 77         | 1.19               | -0.88                          | 0.282                          |
|   | 300        | 1.03               | -0.44                          | 0.110                          |
| 6                                       | 5          | 0.82               | -0.30                          | 0.103                          |
|   | 77         | 0.80               | -0.28                          | 0.097                          |
|   | 300        | 0.59               | -0.12                          | 0.040                          |
| 6.5                                     | 5          | 0.75               | 0.002                          | 0.045                          |
|   | 77         | 0.73               | 0.004                          | 0.042                          |
|   | 300        | 0.50               | 0.000                          | 0.024                          |

The anisotropy field is the minimum field needed to rotate the magnetic moments from the easy to the hard direction. The expressions obtained according this definition are  $H_a = (2K_1 + 4K_2)/\mu_0 M_s$  for easy axis anisotropy, and  $H_a = 2K_1/\mu_0 M_s$  for easy plane anisotropy.

The values of  $K_1$  and  $K_2$  obtained for  $Y_2Fe_{17}$  are in good agreement with those obtained from measurements on aligned samples [15]. The calculated values of  $H_a$  are plotted in figure 2. For  $Y_2Fe_{17}$  and  $Y_2Fe_{16}Ga_1$  our data agree well with those determined by the singular point detection technique [16], except for the value for  $Y_2Fe_{17}$  at 300 K.



**Figure 2.** Concentration dependence of anisotropy field at 5 K (○) and 300 K (●) of  $Y_2Fe_{17-x}Ga_x$ . The data points at 300 K for  $x > 6$  are calculated from the torque data for  $K_1$  and  $K_2$ . The data determined by the singular point detection technique [15] are given by (×) and (+) symbols. The lines are guides for the eye.

### 3.2. Torque curves

It must be noted that, as a consequence of the vanishing anisotropy for the samples with Ga concentrations higher than  $x = 6.5$ , the magnetization curves measured both in the hard and easy direction almost coincide, indicating that the data for the anisotropy constants determined from the magnetization curves are not reliable. In this case torque curves were measured to determine the anisotropy constants at room temperature for compounds with  $H_a$  low enough to be obtainable in our torque magnetometer fields of 1.3 T. The torque, in the plane containing the  $c$ -axis, can be described as

$$L(\theta) = -dE_a/d\theta = -2K_1 \sin \theta \cos \theta - 4K_2 \sin^3 \theta \cos \theta - 6(K_3 + K'_3 \cos 6\phi) \sin^5 \theta \cos \theta + \dots \quad (6)$$

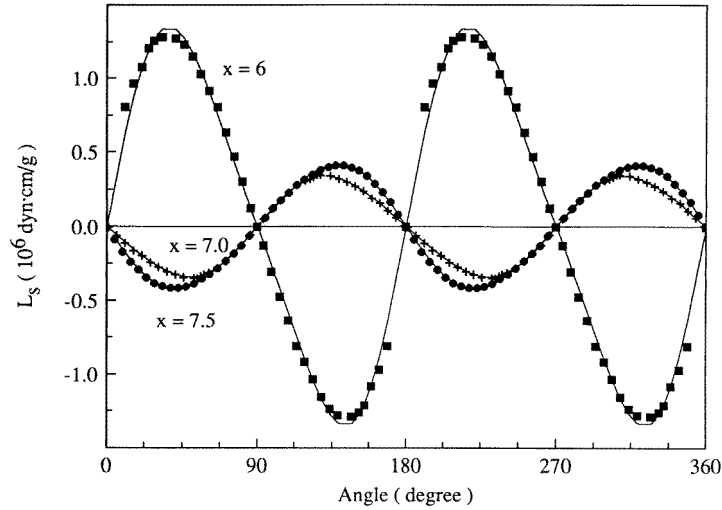
Using a least-squares routine, the Fourier coefficients can be determined from

$$L(\theta) = b_0/2 + b_2 \sin(2\theta) + b_4 \sin(4\theta) + b_6 \sin(6\theta) + \dots \quad (7)$$

The basal plane anisotropy,  $K'_3$  must be determined from a measurement of  $L(\theta)$  in the  $a$ - $b$ -plane on a monocrystal. As the oriented samples under study are isotropic in the plane normal to the  $c$ -axis, it is not possible to obtain  $K_3$  and  $K'_3$  separately. Considering only the first two terms of  $L(\theta)$ ,  $K_1$  and  $K_2$  can be determined from the appropriate relations between the Fourier coefficients and the anisotropy constants:

$$\begin{aligned} K_1 &= -b_2 - 2b_4 \\ K_2 &= 2b_4. \end{aligned} \quad (8)$$

Torque curves were measured in two ways: as a function of  $\theta$  at a constant field and as a function of field at a constant angle. The saturated values of the torque,  $L_s$ , were obtained by extrapolation according to the dependence  $L(H) = L_s(1 - C/H^2)$ , which fits the experimental curves  $L(H)$  at fields higher than 0.8 T. The torque curves



**Figure 3.** Torque curves of  $Y_2Fe_{17-x}Ga_x$ , measured in the plane containing the  $c$ -axis. The torque curves, calculated from the anisotropy constants, using the expression  $L_{calc} = -(K_1 + K_2) \sin 2\theta + (1/2)K_2 \sin 4\theta$ , are shown by the solid line.

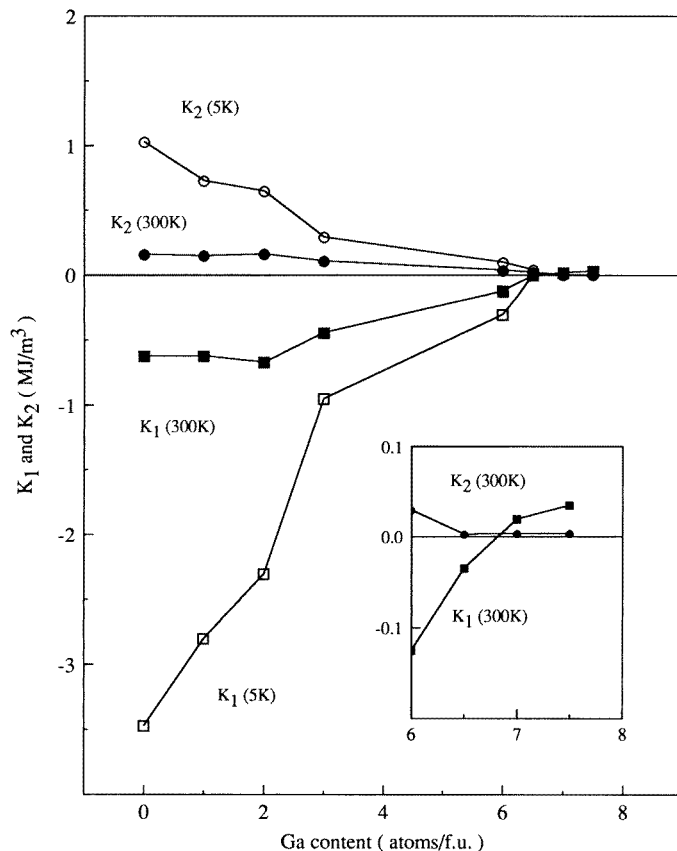
$L$  against  $\theta$  for some of the samples are given in figure 3. The torque curves  $L_s(\theta)$  were analysed by a standard Fourier analysis. The anisotropy constants were calculated using (8). The torque curves, calculated from these constants, using the expression  $L_{calc} = -(K_1 + K_2) \sin 2\theta + (1/2)K_2 \sin 4\theta$ , derived from (6) are shown by the solid line in figure 3. Negligible values of  $K_3$  and  $K'_3$  are inferred by the agreement between the corrected torque curves  $L_s(\theta)$  and  $L_{calc}(\theta)$  (the curves are calculated using the obtained values of  $K_1$  and  $K_2$ ). The error in the anisotropy constants from neglecting the higher-order Fourier coefficients is estimated to be less than 10%. The values obtained for  $K_1$  and  $K_2$  are given in table 2.

**Table 2.** Anisotropy constants of  $Y_2Fe_{17-x}Ga_x$  at room temperature determined by torque measurement.

| Ga content<br>(atoms fu <sup>-1</sup> ) | $K_1$<br>(MJ m <sup>-3</sup> ) | $K_2$<br>(MJ m <sup>-3</sup> ) |
|---|--------------------------------|--------------------------------|
| 6                                       | -0.125                         | 0.030                          |
| 6.5                                     | -0.034                         | 0.003                          |
| 7                                       | 0.020                          | 0.004                          |
| 7.5                                     | 0.035                          | 0.004                          |

### 3.3. $K_1$ and $K_2$ as a function of Ga concentration

The concentration dependence of  $K_1$  and  $K_2$  at various temperatures is shown in figure 4.  $K_1$  at 5 K decreases monotonically becoming less negative with Ga concentration, and thus indicating a weakening of the easy plane character of the Fe anisotropy which is characterized by a negative value of  $K_1$ . The Fe anisotropy becomes uniaxial for Ga slightly above 6.5 atoms fu<sup>-1</sup> at which point  $K_1$  changes sign (see the inset in figure 4).



**Figure 4.** Concentration dependence of the anisotropy constants at 5 and 300 K. The torque data are plotted in the inset. The lines are guides for the eye.

The compound with  $x = 6.5$  has an easy cone type of anisotropy according to the results of x-ray analysis [6] at room temperature. It should be kept in mind that the alignment of such a powder in a static field would produce a sample in which the  $c$ -axes of the individual grains are around the direction of the aligning field, along a cone with half-angle equal to the easy cone angle of the compound. In this case, the obtained values for  $K_1$  and  $K_2$ , and hence for  $H_a$ , have to be regarded as effective values for the sample, different from the intrinsic values for the compound. At room temperatures, figure 4,  $K_1$  does not change noticeably until  $x = 3$ , as a consequence of the rise in  $T_c$  in this region of  $x$  [6], but it decreases with a further increase of Ga before changing sign, as observed at low temperatures. It can be seen from the torque data plotted in the inset of figure 4 that an increase of Ga concentration, above the value at which the change of the anisotropy type takes place, results in a slight increase in  $K_1$ , i.e. in a strengthening of the uniaxial anisotropy, although the decrease of  $T_c$  with  $x$  for this region of Ga concentration [6] has the opposite effect.  $K_2$  is positive and decreases with Ga concentration for all temperatures. It is worth noting that the obtained values of  $K_1$  and  $K_2$  fulfil the conditions required for easy axis and easy plane anisotropy.

It is well known that Fe atoms occupy four non-equivalent sites in the rhombohedral  $Th_2Ni_{17}$ -type structure adopted by  $Y_2Fe_{17-x}Ga_x$  [6] (for  $x = 2$  and 3 the  $Th_2Ni_{17}$  structure



coexists with the  $\text{Th}_2\text{Zn}_{17}$ -type structure). For a compound with different crystallographic sites for the 3d ions, the first-order anisotropy constant  $K_1$  can be expressed [17] by  $K_1 = n_i K_1^i$ , where  $n_i$  is the number of 3d ions in the site  $i$  and  $K_1^i$  is the corresponding contribution of that site to the anisotropy constant  $K_1$ . Such a model has been applied in [18] to  $\text{Y}_2\text{Fe}_{17}$  and  $K_1^i$  have been found to have the values of  $-0.17$ ,  $+0.35$ ,  $-1.14$ , and  $+0.60 \text{ MJ m}^{-3}$  on Fe 18f, 18h, 9d and 6c sites, respectively. Neutron diffraction studies [10] of  $\text{Y}_2\text{Fe}_{17-x}\text{Ga}_x$  ( $R = \text{Nd}$  and  $\text{Tb}$ ) indicate that Ga completely avoids the 9d sites, occupies the 6c ‘dumbbells’ site only at high values of  $x$  and strongly prefers the 18f site at high values of  $x$ . Taking  $K_1^i$  as constants with Ga substituting Fe and considering the change of total  $K_1$  as a result of the absence of Fe atoms, will give an opposite trend of the  $K_{1-x}$  relation to that found experimentally. In fact the replacement of Fe by Ga changes the local anisotropy because the charge distribution around each Fe site and therefore its electric field gradient are different compared to  $\text{Y}_2\text{Fe}_{17}$ . Taking the Fe 9d site as an example, it has four nearest-neighbour Fe 18h atoms in the same  $a$ - $b$ -plane, four Fe 18f atoms in the plane parallel to the  $c$ -axis, and two Fe 6c and two Y 6c atoms out of the  $a$ - $b$ -plane where the Fe 9d atoms are situated. The Fe 9d–18f and Fe 9d–18h distances are quite close to each other ( $2.459 \text{ \AA}$  and  $2.429 \text{ \AA}$ , respectively), as are the bond angles of Fe 18f–9d–18f ( $59.26^\circ$ ) and 18h–9d–18h ( $62.04^\circ$ ). The electric field gradient contributed by the Fe 18f and Fe 18h atoms to the Fe 9d site nearly cancel. The in-plane anisotropy of Fe 9d in  $\text{Y}_2\text{Fe}_{17}$  is mainly due to the out-of-plane 6c atoms. This suggests that Fe 18f will also favour the  $a$ - $b$ -plane but Fe 18h will favour the  $c$ -axis as the easy magnetization directions on Fe 9d sites. When Fe is replaced by Ga, supposing Ga has a negative charge, the opposite is true. At high Ga concentration, the effect of the 18f and 6c sites can surpass that of the 18f site due to their higher Ga occupation and therefore the easy magnetization direction of the Fe 9d site changes from  $a$ - $b$ -plane into  $c$ -axis. The other Fe sites may have similar changes but with less effect because of the absence of Fe atoms on these sites. Therefore, the total anisotropy can change from negative in  $\text{Y}_2\text{Fe}_{17}$  into positive in  $\text{Y}_2\text{Fe}_{17-x}\text{Ga}_x$ . The possible change in the Fe band structure caused by Ga substitution could also be responsible for such a change.

### 3.4. AC susceptibility

For all samples the real component of the ac susceptibility changes smoothly with temperature and no spin reorientation transitions are detected, not even for the sample with  $x = 6.5$  for which such a transition might be expected.

When the field is applied along the hard magnetization direction, the susceptibility is contributed mainly by domain magnetization rotation processes. For easy axis samples

$$\chi_{\perp} = \frac{\mu_0 M_s^2}{2K_1}. \quad (9)$$

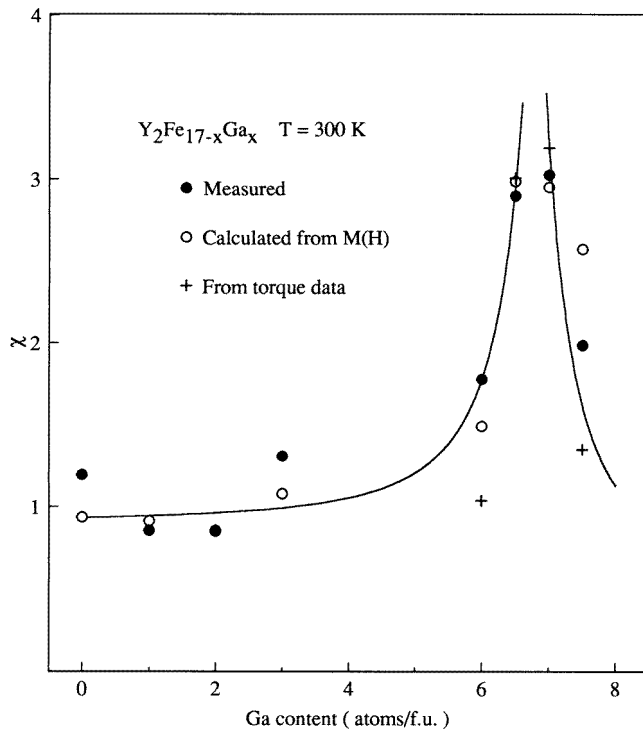
For easy plane samples

$$\chi_{\perp} = \frac{\mu_0 M_s^2}{2K_1 + 4K_2}. \quad (10)$$

If the above two expressions are compared with (2) and (3), used to determine the anisotropy constants, the zero-order parameter from the fit,  $B$ , can be directly related to  $\chi_{\perp}$ .

$$\chi_{\perp} = \frac{1}{B - N}. \quad (11)$$

As for  $K_1$  determination, the result depends on  $N$ . The bigger the value of  $B$  the lower the relative error in  $\chi_{\perp}$ , induced by the demagnetization factor, is.



**Figure 5.** Ac susceptibility at 300 K as a function of Ga concentration. The line is a guide for the eye.

Measured susceptibility (corrected with  $N = 0.3$ ) is compared in figure 5 with the room-temperature susceptibility, calculated according to (9) and (10), and using the data for the anisotropy constants, listed in tables 1 and 2. As expected, the susceptibility initially increases with increasing Ga concentration because of vanishing anisotropy and after exceeding the critical concentration at which a change of anisotropy takes place decreases because of the increase of  $K_1$ .

#### 4. Conclusions

Substitution of Ga for Fe in  $Y_2Fe_{17-x}Ga_x$  initially decreases the anisotropy constants thus indicating a weakening of the easy plane character of the Fe anisotropy. The Fe anisotropy becomes uniaxial for Ga concentration slightly above 6.5 atoms  $fu^{-1}$  at which the first anisotropy constant  $K_1$  changes sign and becomes positive. A strengthening of the uniaxial anisotropy with further increase of Ga concentration is indicated by an increase in  $K_1$ , determined by torque measurements at room temperatures. The results show the important changes which Fe anisotropy undergoes when Fe atoms are substituted by Ga in  $Y_2Fe_{17}$  compounds. The present results can be used to separate the contribution of the two sublattices in  $R_2Fe_{17-x}Ga_x$  compounds where R is a magnetic rare earth, and thus to determine the influence of Ga on the magnetic anisotropy in these compounds.

## Acknowledgments

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