MÖSSBAUER EFFECT STUDY OF SPIN REORIENTATION IN ERBIUM IRON GARNET

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Abstract.- The spin reorientation in erbium iron garnet has been investigated with ^{57}Fe Mössbauer effect. It is shown that the easy magnetic axis rotates in a continuous way in the temperature range 65(5) - 95(5) K from the <100> to the <111> direction through the angle 54°44' in a {110} plane.

1. Introduction.- Much attention has been paid recently to the investigations of magnetic phase transitions of the order-order type in rare-earth magnets, and in particular to the studies of the spin reorientation phase transitions (1). Among different methods used in studies of these transitions, the Mössbauer effect has proved to be a very useful tool (2). The temperature range of spin reorientation in garnets is usually determined from qualitative analysis of the shape of Mössbauer spectra (2,3). The present paper shows that more quantitative information on the spin reorientation in garnets can be derived from studies of temperature dependence of effective quadrupole interaction.

The confusion in the literature concerning the direction of the easy magnetic axis in erbium iron garnet (ErIG) has been discussed recently in (3,4). Other references pertinent to this problem and not cited in (3,4) can also be consulted (5-10). Optical measurements by Belyaeva et al. (5-7) showed that at 4.2 K the easy magnetic axis is in the [100] direction. This was also found by Streever and Caplan with ^{167}Er NMR (8). ^{57}Fe Mössbauer spectra measured below the Néel temperature T_N were analysed without taking into account the orientation of the easy magnetization axis (9,10). In the analysis of ^{166}Er Mössbauer spectra measured in the temperature range 4.2 - 85 K it was assumed incorrectly that the [111] direction is the easy magnetic axis (9). The main cause of the confusion associated with the direction of the easy axis of magnetization in ErIG was the narrow temperature range of most measurements.

2. Analysis and discussion.- ^{57}Fe Mössbauer spectra of ErIG have been measured in the temperature range 1.5 - 579 K. The experimental details have been described elsewhere (3).

It can be shown for a cubic ferrimagnet that when the first two constants of magnetic anisotropy are only taken into account, then a minimum of the free energy is achieved only for the orientation of the magnetization along one of the three different crystallographic directions: [100], [111] or [110] (1). Since the signs and magnitudes of anisotropy constants may change with temperature, this may lead to a reorientation of the magnetization.

The octahedral (a) and tetrahedral (d) sites in rare-earth iron garnets $\{RE_3\}[Fe_2](Fe_3)O_{12}$ are magnetically equivalent or inequivalent depending on whether the angles θ between the direction of magnetization and the axes of local symmetry at the a and d sites are equal or not. The point symmetry $\bar{3}$ of the a sites requires the EFG tensor

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Table 1.- Characteristics of iron sites in $\{RE_3\}[Fe_2](Fe_3)O_{12}$ for three easy directions of magnetization. RI - relative number of iron ions occupying sites characterized by a given angle θ.

Iron site	Easy direction of magnetization								
	[100]			[111]			[110]		
	θ	RI	State	θ	RI	State	θ	RI	State
Octahedral (a)	$54^o44'$	4	a	$70^o32'$	3	a_2	90^o	2	a_2
				0^o	1	a_1	$35^o16'$	2	a_1
Tetrahedral (d)	90^o	4	d_2	$54^o44'$	6	d	45^o	4	d_2
	0^o	2	d_1				90^o	2	d_1

to be axially symmetric and the symmetry axes are equally distributed among the [111] directions. The point symmetry $\bar{4}$ of the d sites also requires an axially symmetric EFG tensor with the axes of symmetry equally distributed among the [100] directions. The values of the angles θ for the three possible easy axes of magnetization in $RE_3Fe_5O_{12}$ are summarized in table 1. Thus, for the easy magnetic axes in the directions [100], [111] or [110], the Mössbauer spectra should consist of three, three or four nuclear Zeeman patterns, respectively, with the intensity ratios RI (table 1).

In the spin reorientation region the Mössbauer spectra of ErIG should be fitted with seven Zeeman patterns (four a-site and three d-site patterns) for an arbitrary direction of the easy magnetic axis. However, since all three possible directions of the easy axes of magnetization lie in {110} type planes it is reasonable to assume that the spin reorientation in ErIG takes place in the {110} type planes. Then the number of d-site Zeeman patterns reduces to two (θ_1 and $\theta_2=\theta_3$ Zeeman patterns - figure 1), and the number of a-site Zeeman patterns reduces to three (the angles α_i between the magnetization and the a-site local symmetry axes are: $\alpha_1 = 54^o44' - \theta_1$, $\alpha_2 = 54^o44' + \theta_1$, $\alpha_3 = \alpha_4 = \cos^{-1}(3^{-\frac{1}{2}}\cos\theta_1)$). In garnets the quadrupole coupling constant $\Delta E_q = \frac{1}{2}e^2qQ$ at the a site is about two times smaller than the one at the d site (11). This causes a considerable overlap of the three a-site Zeeman patterns. Therefore, the spectra in the reorientation region have been fitted with two d-site and one a-site Zeeman patterns. The distance $\Delta v_i(d) = v_i(d_2) - v_i(d_1)$, $i = 1,2...6$, between the i-th d_2 and d_1 lines is equal to

$$\Delta v_i(d) = \frac{3}{4}\Delta E_q(d)(\cos^2\theta_1 - \cos^2\theta_2). \qquad (2.1)$$

From figure 1 one can find that

$$\cos^2\theta_2 = \frac{1}{2}\sin^2\theta_1. \qquad (2.2)$$

This leads to

$$\Delta v_i(d) = \frac{3}{8}\Delta E_q(d)(3\cos^2\theta_1 - 1). \qquad (2.3)$$

The value of $\Delta E_q(d)$ can be obtained from the fit of Mössbauer spectra measured at helium temperatures (3). By measuring the distance $\Delta v_i(d)$ in the spin reorientation region one is able to determine from equation (2.3) the temperature dependence of the angle θ_1, and consequently the temperature range of spin reorientation.

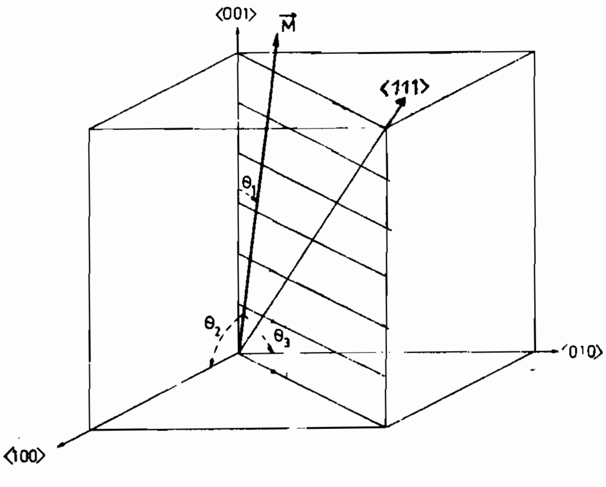

Fig. 1. Geometry of the spin reorientation in a {110} plane.

Mössbauer spectra between 1.5 and 60 K are characteristic for the [100] direction of the magnetization (figure 2). In this temperature range the values of $\Delta v_i(d)$ remain constant within the experimental error. The spectra at 99.5 K and higher temperatures show that the [111] direction is the easy magnetic axis. In the spin reorientation region $\Delta v_i(d)$ decreases with temperature (figure 2). This corresponds to the increase of θ_1 with temperature (figure 3). Figure 3 shows that the spin reorientation occurs gradually over a wide temperature region of about 30 K. The temperatures of an onset and completion of the spin reorientation are 65(5) K and 95(5) K, respectively. It is concluded from the variation of the angle θ_1 as a function of temperature that the easy magnetic axis in ErIG rotates in a continuous way in the temperature range 65(5) - 95(5) K

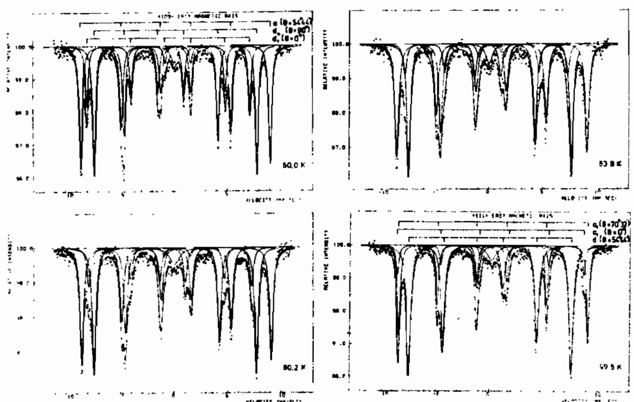

Fig. 2. Representative temperature spectra of ErIG. The solid lines are the best fitted theoretical spectra with their Zeeman components. A broad single line near zero velocity comes from Fe impurities in the absorber holder.

from the <100> to the <111> direction through the angle $54^{\circ}44'$ in a {110} plane. In recent papers by Guillot et al. (12,13) it has been found from magnetization measurements that the [100] direction is the easy one below 50 K (12), whereas specific heat measurements gave the value 54(2) K (13). The spin reorientation process in garnets involves the changes of magnetic space groups. This problem has been discussed in (13,14).

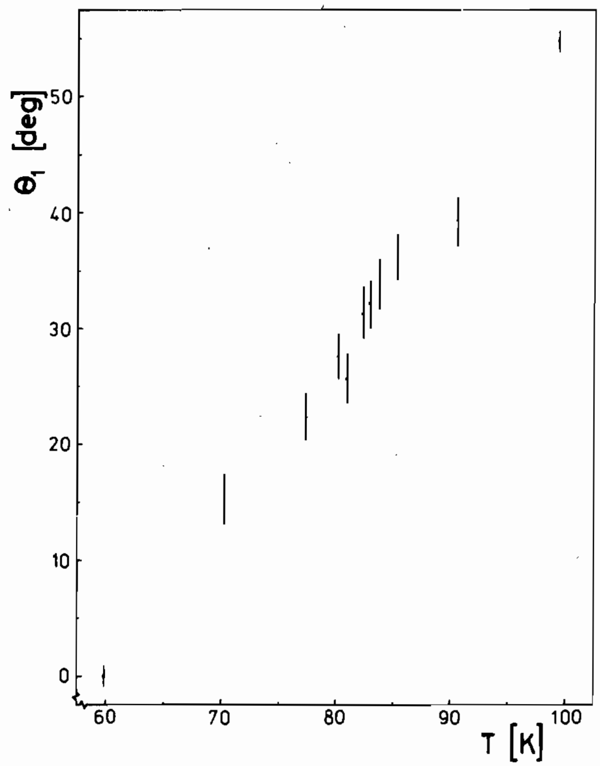

Fig. 3. Temperature dependence of the angle θ_1.

Since below 65(5) K the easy axis is in the [100] direction and above 95(5) K - in the [111] direction, this offers a unique possibility of determining the signs of ΔE_q at both a and d sites. They are both negative in ErIG (3). The negative signs of ΔE_q at those sites were also found in TbIG and YIG (15). This is in accordance with monopole point-charge lattice calculations which show that in all REIG the signs of ΔE_q are negative at both sites (16). The only exception is SmIG where the signs of $\Delta E_q(a)$ and $\Delta E_q(d)$ are negative and positive, respectively (17,18).

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