On the Mechanism of Spin Reorientation in YIG:Si*

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The magnetocrystalline anisotropy theory developed earlier by us and based on a novel energy level model for Fe^{2+} in YIG:Si(Ge) is used here to study the mechanism of spin reorientation in YIG:Si. Single-ion anisotropy with a uniform distribution of Fe^{2+} ions indicate a broad spin reorientation which turns out to be due to the higher-order cubic terms K_3 and K_4. It is found that the completion temperature (T_2) of the spin reorientation is strongly influenced by K_4 thus indicating an inadequacy of the approach based on K_1, K_2 and K_3 only. From the single Fe^{2+} ion anisotropy results we propose a two-center model which implies a new mechanism of the spin reorientation in YIG:Si. The bulk onset (T_1') and completion (T_2') temperatures are linked to the relative number ($\boldsymbol{a}$) of the near Fe^{2+} sites. Spin orientation diagrams in the plane $\boldsymbol{a}-T$ are established for the total $Fe^{2+}-Fe^{3+}$ system for a wide range of the parameters involved. The two-center model accounts well for the bulk experimental temperatures T_1' and T_2' in YIG:Si and for the absence of spin reorientation in samples having Si-content less than 0.18. The total magnetization $\boldsymbol{M}$ is predicted not to rotate smoothly between the [100] and [111] direction. There exists an upper critical angle beyond which $\boldsymbol{M}$ switches sharply to [111] as the temperature approaches T_2'. Comparison with the previous model which requires a nonuniform distribution of Fe^{2+} ions is also discussed.

1. Introduction

During the past few years, the studies of spin reorientations in rare-earth cubic magnets have attracted growing interest [1–6]. Recently, composition dependent spin reorientations have been reported for $Y_3Fe_{5-x}Si_xO_{12}$ [7]. The easy magnetic axis in YIG:Si rotates from the [111] direction at high temperatures to the [100] direction at low temperatures. Although it is certain that the induced Fe^{2+} ions are responsible for the continuous transition in YIG:Si, its mechanism is not well understood. A similar spin transition has quite recently been observed also in YIG (Ru, Ge) and YIG (Co, Ge) [8].

It is well known [1, 9, 10] that the magnetic cubic anisotropy (in the usual notation)

$$F_a = K_1\Phi + K_2 Q + K_3\Phi^2 + K_4\Phi Q + \ldots \qquad (1)$$

truncated to the first two terms in (1) can account only for abrupt spin reorientations between the three major directions [100], [110] and [111]. The significance of the third term in (1) for continuous spin reorientations has only recently been recognized [11, 12] in the studies on RE-iron Laves compounds [2]. However, in the studies on YIG:Me^{4+} (Me = Si or Ge), until recently, only K_1 and K_2 have been considered and the anisotropic ground doublet energy level [13] has been widely applied for the induced Fe^{2+} ions. The orbital singlet ($^5A_{1g}$) ground state model was found unsatisfactory [14, 15].

In order to account for the continuous nature of the transitions in YIG:Si within the framework of the only doublet model available at that time [13] it was necessary to assume [7] a nonuniform distribution of Fe^{2+} ions (see Section 2.1) in the whole temperature range velow T_c. However, the nonuniform distribution is generally expected rather at low temperatures only, while due to thermal agitation a uniform distribution is more likely at higher temperatures. Besides, the cubic anisotropy studies on YIG:Si(Ge) [16–19] were based on the uniform distribution of the Fe^{2+} ions. The temperature-independent imbalance in the site distribution [7] implies an intrinsic unaxial anisotropy also at room temperatures. To the best of our knowledge a weak uniaxial anisotropy was observed in YIG:Si only

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below 30 K [16]. Neither that aspect nor cubic anisotropy has been explicitely considered in [7].

In our papers [20, 21], simultaneous with the discovery [7], we have investigated the effect of a nontrigonal crystal field on the spectroscopic properties of Fe^{2+} ions in YIG : Si (Ge). The studies [20, 21] have provided a novel energy level model for the Fe^{2+} ions based on an orbital singlet ground state different from the $^5A_{1g}$ singlet [15]. Consequently we have developed the magnetocrystalline anisotropy theory for Fe^{2+} ions in YIG : Me^{4+}. The constants K_1 and K_2 have been derived explicitly at 0 K [22]. The temperature dependence of the magnetic anisotropy has revealed [23] a pronounced role of the higher-order anisotropy terms K_3 and K_4 for the present model [21] as well as for the previous one [13, 19]. The magnetic anisotropy theory based on the spin Hamiltonian [21] for $3d^6(3d^4)$ ions at triclinic symmetry sites has been published in full elsewhere [24]. The results [24] suggest our energy level model for Fe^{2+} [21] can account for the broad spin reorientations observed in YIG : Si [7] within the assumption of uniform distribution of Fe^{2+} ions.

It is the purpose of the present paper to study the bearings of our model [21, 24] on the spin reorientations in YIG : Si in more detail. The bearings of the single-ion anisotropy on spin reorientations in the Fe^{2+} subsystem are considered in Section 2. The single-ion results are compared with the corresponding results of the previous model and with the experimental data. Based on this analysis we propose in Sect. 3 a more realistic two-center model [25–27]. Spin orientation diagrams for the total $Fe^{2+}-Fe^{3+}$ system are then worked out for a wide range of the parameters involved. This study elucidates an alternative mechanism of spin reorientations in YIG : Si. In the Appendix we list the experimental K_1 and K_2 for the pure YIG [9, 28] used in our calculations.

2. Single-ion Anisotropy

2.1. Preliminary calculations

The Fe^{3+} ions in pure YIG give negative K_1 and K_2 [9, 28] (see Appendix) and hence the minimum anisotropy energy in the [111] direction at all temperatures. In order to explain the spin reorientations in YIG : Si one has to consider a variation of the minimum anisotropy energy of Fe^{2+} ions with temperature [7]. Then the easy magnetization direction for the Fe^{2+} subsystem as well as for the total $Fe^{2+}-Fe^{3+}$ system can be determined.

The simplest approach to cope with the large variety of crystallographically different Fe^{2+} centers [25–27] which are, in fact, induced in YIG : Me^{4+} is to assume one kind of the octahedral Fe^{2+} centers. Then the Fe^{2+} sites may differ only in the relative orientation of the local axis system. The distribution of Fe^{2+} ions over the four [19, 7] or twelve [29] orientationally inequivalent sites may be either uniform or nonuniform. As the previous model [13] allows hardly to distinguish between the crystallographically different Fe^{2+} centers its applications have never gone beyond the one-center approach. Our model [20, 21] is not subject to this constraint [22] (see Section 3), however, we consider first this approach to allow for comparison with the previous model.

The free energy F_t of a Fe^{2+} ion with the framework of our model [21, 24] depends on the eight spin Hamiltonian parameters $B_0^{(2)}$, $B_1^{(2)}$, $B_2^{(2)}$, $B_0^{(4)}$, $B_1^{(4)}$, $B_2^{(4)}$, $B_3^{(4)}$ and $B_4^{(4)}$, the molecular field $\boldsymbol{h} = g_B \boldsymbol{H}_{ex}$, and the site occupancies n_i. Because energy-level calculations [24] indicate that the nonuniform distribution is not favoured by the present model we consider here only the uniform distribution of Fe^{2+} ions over the twelve [29] orientationally inequivalent sites, i.e. $n_i \equiv 1$, for $i = 1-12$. Using the equations derived earlier [24] the anisotropy energy of a Fe^{2+} ion is calculated as a function of the angle δ between the magnetization $\boldsymbol{M}$ and the [100] direction in the plane $(01\bar{1})$, we have

$$F_a^t(\delta_q) = F_t(\delta_q) - F_t(\delta = 0°) . \tag{2}$$

The angle δ is varied in steps 5° between 0° ([100]) and 50° and with the last $\delta = 54.74°$ ([111]); thus $q = 1-12$ in (2). Considering a set of the $F_a^t(\delta_q)$ values, $q = 1, \ldots, 12$, for a given set of the $B_q^{(k)}$'s and $\boldsymbol{h}$ at a certain temperature we find a minimum anisotropy energy and the respective angle δ_q. There are three different cases [24]: (a) the minimum of $\{F_a^t(\delta_q)\}$ is at $\delta_1 = 0°$, i.e. $\boldsymbol{M}$ [100] at all T between 0 and 300 K, (b) the minimum is at $\delta_{12} = 54.74°$, i.e. $\boldsymbol{M}$ [111] at all T, and (c) the minimum gradually shifs from the [100] direction at low T towards the [111] direction at high T. Table 1 illustrates the results [24] for the $B_q^{(k)}$ sets predicted by the microscopic theory [21] and several $\boldsymbol{h}$ values in the range 150 to 500 cm^{-1} suitable for YIG [9, 10,

Table 1. General features (see text) of the anisotropy energy of Fe^{2+} ion in YIG:Si within the uniform distribution model for the various $(B_q^{(k)}, \boldsymbol{h})$ sets. The nontrigonal (Γ) and trigonal (Δ) crystal field parameters and the molecular field $\boldsymbol{h}$ are in cm^{-1}.

Γ	200	200	200	200	300	300	300	300	300
Δ	300	400	500	600	300	400	500	600	700
$B_q^{(k)}$ set	1	2	3	4	5	6	7	8	9
$h = 200$	c	a	a	a	b	b	a	a	a
300	a	a	a	a	b	b	a	a	a
400	a	a	a	a	b	b	a	a	a

16–19]. The case (c), i.e. a broad spin reorientation has also been found for some other $(B_q^{(k)}, \boldsymbol{h})$ sets [24]. One may expect that for some $B_q^{(k)}$ values intermediate between the set no. 6 and 7 the anisotropy energy of a Fe^{2+} ion would also exhibit the property (c). Subsequent numerical calculations prove this is actually the case.

One point should be mentioned. The theory with K_1 and K_2 in (1) only predicts the easy magnetization direction along one of the three major directions (see Sect. 1) depending on the sign and relative magnitude of K_1 and K_2 [1, 9, 10]. Although these rules do not apply if the higher-order terms in (1) are taken into account it is worthwhile to calculate also F_a^t for the direction [110] to be sure that it does not fall below the minimum of $F_a^t(\delta_q)$. In no case considered in Table 1 and in the subsequent calculations F_a^t [110] is lower than the corresponding minimum of $F_a^t(\delta_q)$. Hence, the direction [110] is not an easy magnetization direction.

The previous model [13] with a variable distribution of Fe^{2+} ions approaching a uniform distribution with decreasing temperature indicated only abrupt spin reorientations [7]. In order to check the behaviour of that model with a pure uniform distribution we have computed $F_a^t(\delta_q)$ and F_a^t [110] with the three different values of λ and f [17–19]. The calculations yield (i) the minimum anisotropy energy exclusively in the direction [100] and (ii) significant K_3 and K_4 at low temperatures [23, 24]. The higher-order anisotropy terms have been neglected in all previous studies [9, 14, 16–19] on YIG : Me^{4+}.

2.2. *Spin reorientations in the Fe^{2+} subsystem*

Proceeding in the way described in Sect. 2.1 we have made extensive calculations to find the $(B_q^{(k)}, \boldsymbol{h})$ sets exhibiting a broad spin reorientation. The mechanism of the transition in the Fe^{2+} subsystem will be illustrated on two examples, namely, the $B_q^{(k)}$ set (i) equivalent with the set 1 in Table 1 and the set (v) intermediate between the set 6 and 7, i.e. with $B_0^{(2)} = 4.8$, $B_1^{(2)} = -2.5$, $B_2^{(2)} = -1.3$, $B_0^{(4)} = 0.0060$, $B_1^{(4)} = -0.0155$, $B_2^{(4)} = 0.0015$, $B_3^{(4)} = 0.0001$ and $B_4^{(4)} = 0.0004\ cm^{-1}$. The anisotropy energy $F_a^t(\delta)$ is plotted vs. the angle δ in Fig. 1 for several temperatures in the spin reorientation region. The variation of the position of the minimum anisotropy energy with temperature is depicted in Fig. 2 for the set (v) with several $\boldsymbol{h}$ values.

It is seen in Fig. 2 that the set (v) with $\boldsymbol{h} = 160$ and $180\ cm^{-1}$, besides a broad spin reorientation at

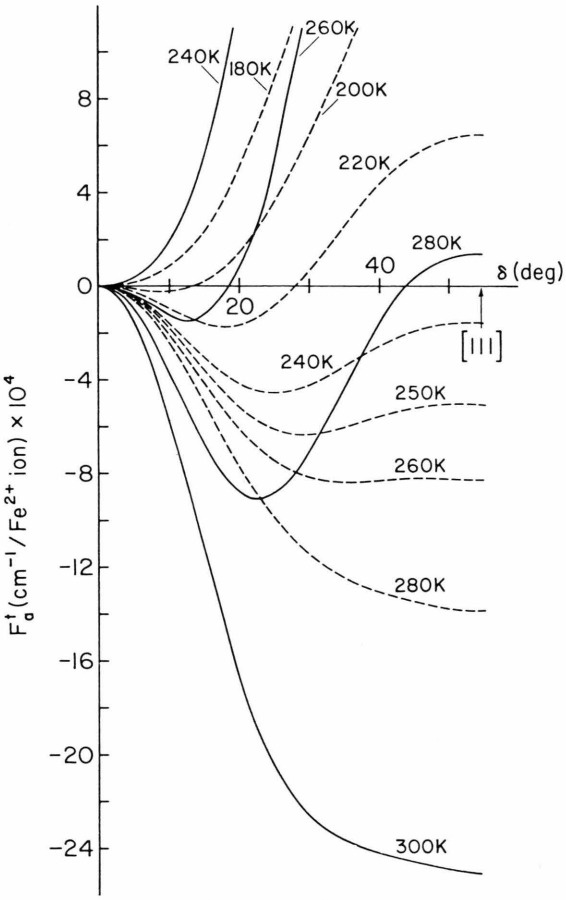

Fig. 1. Anisotropy energy of Fe^{2+} ion in YIG:Si vs. the angle δ in the spin reorientation region for the $B_q^{(k)}$ set (i) [full line] and (v) [broken line] ($\boldsymbol{h} = 200\ cm^{-1}$).

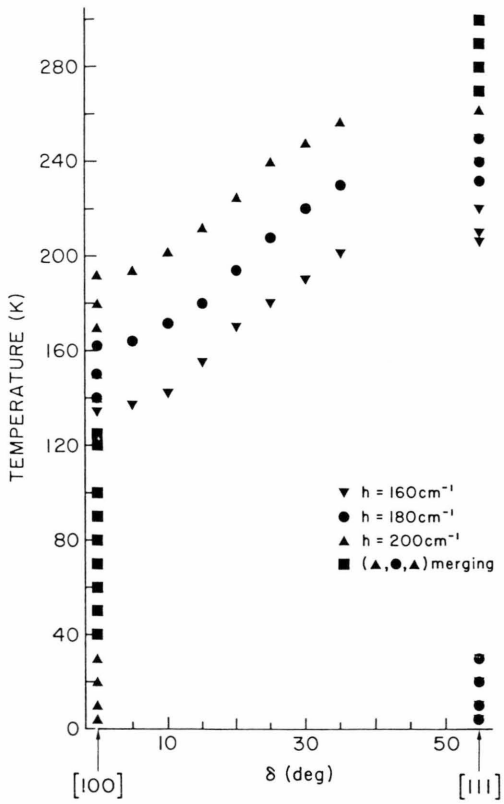

Fig. 2. Positions of the minimum anisotropy energy of Fe^{2+} ion in YIG:Si vs. temperature with the $B_q^{(k)}$ set (v) and various molecular fields.

higher temperatures, also exhibits an abrupt transition at around 30 K where the easy magnetization direction changes from [111] to [100]. No such effect is found with $\boldsymbol{h} = 200\ \mathrm{cm}^{-1}$ and above.

Figures 1 and 2 reveal another interesting feature of the broad spin reorientations in the Fe^{2+} subsystem in YIG:Si. Let us denote by T_1 the onset and by T_2 the completion temperature of the transition with increasing T. The angle δ_m at which the minimum anisotropy energy occurs at a given T does not vary smoothly between 0° and 54.74°. There exists certain upper critical angle δ_{mc} beyond which $\boldsymbol{M}$ rotates abruptly within a very narrow temperature range into the direction [111] as T approaches from below T_2. For the set (v) with $\boldsymbol{h} = 160-200\ \mathrm{cm}^{-1}$ the upper critical angle δ_{mc} is about 35°. It is seen in Fig. 1 that for T close to T_2 the anisotropy energy $F_a^t(\delta)$ has no stable minimum and only a smeared minimum region between δ_{mc} and 54.74°. This intrinsic feature of the anisotropy energy seems to be quite novel in view of the up-to-now studies on the spin reorientation processes.

The analytical expressions [24] for the constants K_1, K_2, K_3 and K_4 in terms of free energy $F_t[hkl]$ for some five chosen $[hkl]$ directions have been used to study the temperature dependence of the anisotropy (in Eq. (9) of [24] K_4 should read $-K_4$). The results for one chosen $(B_q^{(k)}, \boldsymbol{h})$ set are shown in Figure 3. The temperature ranges in which the term K_3 and K_4 is greater than 10% [30] of the term K_1 are indicated by a heavy line in Fig. 3 to visualize the significance of the higher-order terms [24]. The ground energy in our model does not contain, in the approximation used [22], any terms which may contribute to K_3 and K_4. Therefore our K_3 and K_4 must be zero at 0 K, while due to the population of the excited states [24] they become nonzero at finite

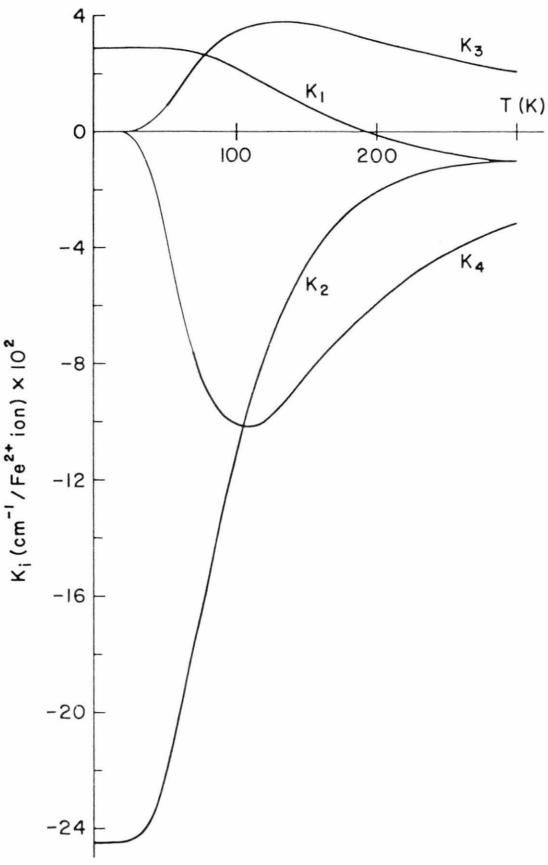

Fig. 3. Temperature dependence of the cubic anisotropy constants for Fe^{2+} ion in YIG:Si with the $B_q^{(k)}$ set (v) and $\boldsymbol{h} = 200\ \mathrm{cm}^{-1}$.

temperatures. As we focus this paper on the mechanism of spin reorientation the diagrams $K_i(T)$ for the other $(B_q^{(k)}, \boldsymbol{h})$ sets will not be presented here. It is enough to say that in all cases where a broad spin reorientation is found the constants K_3 and K_4 are significant over a wide temperature range. Hence, in view of the predictions [1, 9, 10] based on K_1 and K_2 as well as those [11, 12] based on K_1^{I}, and K_2^{I} and K_3^{I} [23] (see below) our results show clearly that these are the higher-order cubic anisotropy terms which are responsible for the continuous nature of the transitions indicated by the present model.

The constants [12] K_i^{I} are related to the K_i's [3, 10, 24] in (1) as follows [23]:

$$K_1^{\mathrm{I}} = K_1\,, \quad K_2^{\mathrm{I}} = K_2 + 2K_3\,, \quad K_3^{\mathrm{I}} = K_3\,. \tag{3}$$

Hence, in terms of the K_i's the condition [12] for the lower boundary of spin reorientation is given by $K_1 = 0$, $K_3 > 0$. A comparison of Figs. 2 and 3 and an inspection of the corresponding data for the other $(B_q^{(k)}, \boldsymbol{h})$ sets indicates, in fact, that (i) the onset temperature T_1 coincides with the temperature at which K_1 changes sign and (ii) K_3 is always positive. Similarly the condition [12] for the upper boundary of the transition can be written as

$$L \equiv K_1/K_3 = -[(K_2/K_3) + 4]^2/24 \equiv R\,, \tag{4}$$

and the completion temperature T_2 can be obtained solving (4), i.e. $L(T) = R(T)$. Using the K_i values for some $(B_q^{(k)}, \boldsymbol{h})$ sets we have found no reasonable solution of (4). Generally, T_2 from (4) was much higher than T_2 found graphically (see, e.g. Fig. 2) or no solution existed below 320 K. This suggests that the presence of K_4 strongly modifies the upper spin reorientation boundary. Hence, an extension of the phenomenological predictions [12] taking into account also K_4 in (1) would be highly desirable.

The mechanism of broad spin reorientations for the other $(B_q^{(k)}, \boldsymbol{h})$ sets is to a great extent similar to that for the set (i) and (v). The relevant results are summarized in Table 2. The sets (ii) and (iii) are two intermediate sets between the set no. 1, i.e. (i) and no. 2, i.e. (iv) (see Table 1). The parameters $B_2^{(2)}$, $B_0^{(4)}$ to $B_4^{(4)}$ are equal to -1.4, 0.026, -0.045, 0.0039, 0.0007, 0.011 and -1.3, 0.037, -0.050, 0.0038, 0.0005, 0.0010 for the set (ii) and (iii), respectively, whereas $B_1^{(2)} = -3.4$ for both sets. For the set (iv) with $\boldsymbol{h} = 160$ and 180 cm^{-1} an abrupt or very narrow spin reorientation occurs at about 250 and 286 K, respectively, whereas with $\boldsymbol{h} = 200\ \mathrm{cm}^{-1}$ no spin reorientation is found below 320 K. The $B_q^{(k)}$ parameters for the set (vi) are identical with those for the complementary set (v), except for the $B_0^{(2)}$, as indicated in Table 2. The set (vii) and (viii) have been found earlier [24] to yield reasonable agreement between our theoretical K_1 and K_2 and the experimental data at low temperatures for Fe^{2+} in YIG:Si and YIG:Ge, respectively. The parameters $B_1^{(2)}$, $B_2^{(2)}$, $B_0^{(4)}$ to $B_4^{(4)}$ are -2.5, -1.7, 0.0160, -0.0170, 0.0015, 0.0, 0.0003 and -3.4, -2.7, 0.1100, -0.0400, 0.0036, 0.0001, 0.0005 for the set (vii) and (viii), respectively.

Table 2. The onset (T_1) and the completion (T_2) temperature of the broad spin reorientation for Fe^{2+} in YIG:Si with various $(B_q^{(k)}, \boldsymbol{h})$ sets (see text) within the uniform distribution model. T in K, $\boldsymbol{h}$ and $B_0^{(2)}$ in cm^{-1}.

$B_q^{(k)}$ set		i	ii	iii	v	vi	vii	viii
$B_0^{(2)}$		6.6	7.3	8.0	4.8	4.9	5.9	10.5
$h = 160$	T_1	180	222	238	134	165	212	232
	T_2	235	255	260	206	235	255	270
$h = 180$	T_1	214	258	274	162	198	248	270
	T_2	265	290	295	232	255	290	300
$h = 200$	T_1	248	294	310	192	230	286	308
	T_2	295	320	–	262	285	320	–

The above studies reveal that the spin reorientation is very sensitive to changes in the spin Hamiltonian parameters and to a lesser extent in the molecular field. The former ones are subject in turn to the changes in the local crystal field around a particular Fe^{2+} site [21]. Although in our calculations we assume the molecular field to be constant in the temperature range 0–300 K, which seems to be quite a reasonable approximation [24], Table 2 also gives an idea about the possible variation of the spin reorientation transition temperatures with the temperature-induced changes in the molecular field.

2.3 Discussion

Both models [13, 21] indicate broad spin reorientations in the Fe^{2+} subsystem [7, 24]. However, the doublet model [13] requires the temperature-independent imbalance in the Fe^{2+} sites distribution [7]. In the singlet model [21] the uniform distribution of Fe^{2+} ions over the octahedral sites, which is well supported by the existing literature [9, 13–19], is sufficient [24]. Moreover, the present model predicts

the magnetization in the Fe^{2+} subsystem outside the spin reorientation region is exactly parallel to the [100] and [111] directions while it is never achieved within the previous model [13] with a nonuniform distribution [7].

Both models allow an interpretation of the composition dependence of the spin reorientations and an explanation of the dissappearance of the transition for specimens having the Si^{4+} content less than 0.18 per formula unit. Model [13] implies [7] an increase in the imbalance with decreasing Si-content. The results in Tables 1 and 2 indicate that even very small changes in the spin Hamiltonian parameter $B_0^{(2)}$ shift significantly the transition region. Our model implies that in the specimens with low Si-content the Fe^{2+} ions have lower Δ [21] (as, e.g. for the set no. 5 or 6) and thus the easy magnetization direction is along [111] at all temperatures and no transition occurs.

The existing experimental data [7] on the transition temperatures in YIG:Si are collected in Table 3. The model [13] does not allow for a clear determination of the transition temperatures T_1 and T_2 for the Fe^{2+} subsystem (see Fig. 5 of [7]). The free energy of the Fe^{3+} subsystem and that of the total $Fe^{2+}-Fe^{3+}$ system have not been quantitatively considered in [7]. As it is seen from Tables 2 and 3 our T_1 and T_2 are close to the experimental values T_1' and T_2', although the interval $(T_2 - T_1)$ is smaller than $(T_2' - T_1')$. For no conclusion on the Fe^{2+} subsystem can be straightforwardly extended to the total $Fe^{2+}-Fe^{3+}$ system the later "discrepancy" cannot be considered as a drawback of the present model. Instead, this "discrepancy" has led us to the idea of a more elaborated model which is presented below.

Table 3. The experimental onset (T_1') and completion (T_2') temperatures of the spin reorientation in $Y_3Fe_{5-x}Si_xO_{12}$ (in K); x_e is the effective Fe^{2+} content per formula unit as found from chemical analysis (after Geller et al. [7]).

x (Si^{4+})	$\leqq 0.18$	0.20	0.25	0.30	0.40
x_e (Fe^{2+})	$\leqq 0.12$	0.13	0.16	0.19	0.25
T_1'	no	?	30	80	90
T_2'	SR	90	215	260	250

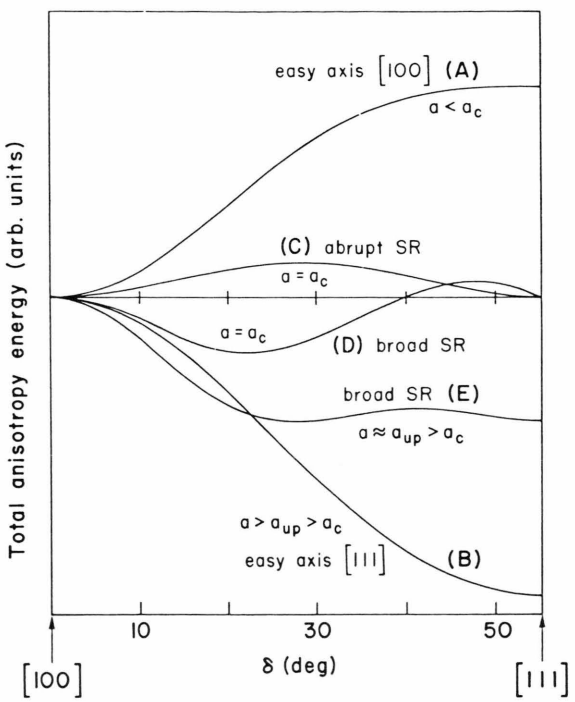

Fig. 4. Total anisotropy energy for YIG:Si in the vicinity of an abrupt (A-C-B) and a broad (A-D-E-B) spin reorientation in the present two-center model. Different scale applies to various curves.

3. Two-center model

3.1. Outline of the model

The qualitative two-center model [25, 26] assumes an existence of two clearly distinguishable Fe^{2+} centers in YIG:Me^{4+}: (i) a "near" center being close to the inducing Me^{4+} ion and (ii) a "far" center located at much larger distance from the Me^{4+} ion. A local distortion around a near center must obviously be stronger than that around a far center. The near Fe^{2+} sites have lower energy and are thought to be preferentially occupied at low temperatures [25–27, 31–33]. The present energy level model unlike the previous one [13] provides clear criteria to distinguish between at least two kinds of Fe^{2+} centers [21, 22]. Thus it enables a quantitative interpretation of the two-centre model [25, 26] in terms of the nontrigonal (Γ) and trigonal (Δ) crystal field parameter [21]. As it has been shown earlier [22] the experimental finding [31] that the near centers are less anisotropic than the far centres is well accounted by our model.

Basing on the strength of a local distortion around Fe^{2+} ion one may consider the $B_q^{(k)}$ sets (v)–(vii) as

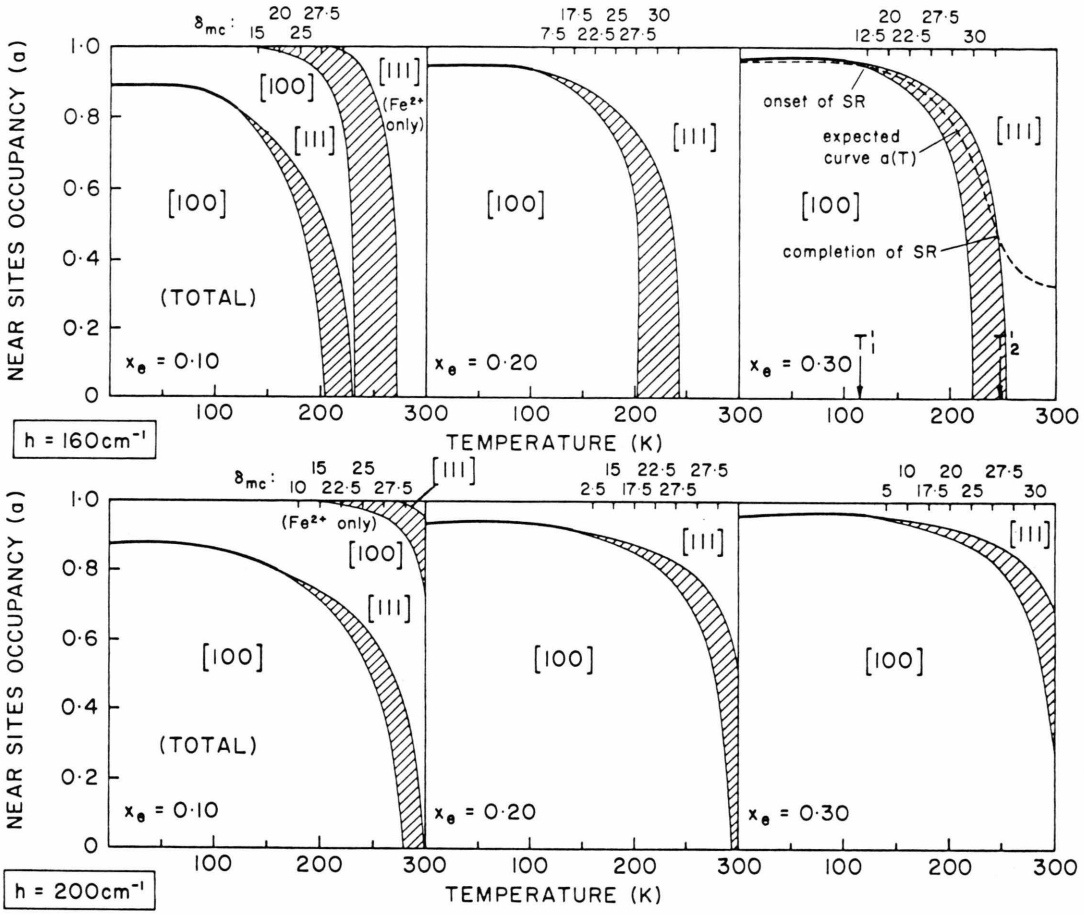

Fig. 5. Spin orientation diagrams in the two-center model for YIG:Si with the $B_q^{(k)}$ set (v) and (viii) (see Table 2) taken for the near and far Fe^{2+} sites, respectively, and with two values of the molecular field ($\boldsymbol{h}$ – in cm^{-1}) and various effective content (x_e) of Fe^{2+} ions. Heavy line – the abrupt spin reorientation boundary, shaded area – the broad spin reorientation region. For explanation of the angle δ_{mc} and the curve $\boldsymbol{a}(T)$ – see text.

appropriate for the near sites, while the other sets in Table 2 may be prescribed to the far sites. If the majority of Fe^{2+} ions occupy the near sites at low temperatures the onset temperature T_1' in the bulk YIG:Si will be mainly influenced by these sites. As with increasing temperature the number of the far sites increases, the bulk completion temperature T_2' will be determined by the relative competition of the near and far sites. A very suitable property of the set (v) is that it yields the lowest T_1 being close to T_1' for YIG:Si with high Si content. On the other hand the sets (ii), (iii) and (viii) yield T_2 close to T_2' (cf. Tables 2 and 3). The above single-ion results has prompted our calculations in the framework of the two-centre model.

Let us denote the relative occupancy of the near and far sites by $\boldsymbol{a}$ and $\boldsymbol{b}$, respectively ($\boldsymbol{a}+\boldsymbol{b}=1$). These quantities must be both temperature and composition dependant. The total anisotropy energy of the $Fe^{2+}-Fe^{3+}$ system is thus given by

$$F_{\mathrm{tot}} = F_{\mathrm{a}}(\mathrm{Fe}^{3+}) + \boldsymbol{a}\, F_{\mathrm{a}}^{\mathrm{n}}(\mathrm{Fe}^{2+}) + (1-\boldsymbol{a})\, F_{\mathrm{a}}^{\mathrm{f}}(\mathrm{Fe}^{2+}), \quad (5)$$

where the superscript $\boldsymbol{n}$ and $\boldsymbol{f}$ refers to the near and far Fe^{2+} sites, respectively. Instead of calculating each anisotropy energy in (5) we use the anisotropy constants. From (1) and (5) we have

$$F_{\mathrm{tot}}(\delta) = \sum_{i=1}^{4} K_i^{\mathrm{tot}} f_i(\delta), \quad (6)$$

where $f_i(\delta)$ are the angle-dependent functions [23, 24] and

$$K_i^{\mathrm{tot}}(\mathrm{erg/cm^3}) = K_i(\mathrm{Fe^{3+}}) + \boldsymbol{a}\bar{K}_i^{\mathrm{n}}(\mathrm{Fe^{2+}}) + (1-\boldsymbol{a})\,\bar{K}_i^{\mathrm{f}}(\mathrm{Fe^{2+}}). \quad (7)$$

We adopt the following conversion factor [28]:

$$\bar{K}_i^{\mathrm{p}}(\mathrm{erg/cm^3}) = x_{\mathrm{e}}\,0.84 * 10^6\,K_i^{\mathrm{p}}(\mathrm{cm^{-1}/Fe^{2+}\ ion}), \quad (8)$$

where x_{e} is the effective Fe^{2+} content per formula unit, $\boldsymbol{p} = \boldsymbol{f}$ or $\boldsymbol{n}$, and K_i^{p} are the single-ion anisotropy constants calculated in Sect. 2.2 and [24]. We assume here that the anisotropy of Fe^{3+} ions in YIG : Si is not affected by Si-doping and consequently we use for $K_i(\mathrm{Fe^{3+}})$ in (7) the experimental data for pure YIG [9, 28] listed in Appendix. The constants K_3 and K_4 for Fe^{3+} ions are taken to be equal zero, which is quite justified for pure YIG [9, 34, 35].

3.2. *Spin orientation diagrams*

The anisotropy energy F_{tot} of (5) is finally a function of the following parameters: δ, T, x_{e}, $\boldsymbol{a}$, and the values $\{K_i^{\mathrm{n}}\} \equiv \boldsymbol{n}$ and $\{K_i^{\mathrm{f}}\} \equiv \boldsymbol{f}$ $(i = 1, \ldots, 4)$. Considering for a given set $(\boldsymbol{n}, \boldsymbol{f})$ F_{tot} vs δ at a constant T and x_{e} the position of the minimum total anisotropy energy can be found as a function of $\boldsymbol{a}$. Then, a spin orientation diagram [11, 12] in the plane $\boldsymbol{a} - T$ can be constructed. In these calculations δ is varied in steps 2.5° between 0° and 52.5° and with the last $\delta = 54.74°$ whereas x_{e} in steps 0.05 between 0.05 and 0.30 (see Table 3).

Let us first discuss the general properties of F_{tot}. The variation of $F_{\mathrm{tot}}(\delta)$ in the vicinity of a transition is depicted in Figure 4. There exists a critical value of $\boldsymbol{a}$ for which F_{tot} (54.74°) is equal zero. With $\boldsymbol{a} = \boldsymbol{a}_{\mathrm{c}}$ the minimum of $F_{\mathrm{tot}}(\delta)$ is either at $\delta = 0°$ and 54.74° for an abrupt transition (curve C in Fig. 4) or at an intermediate δ for a broad transition (curve D). In the later case the upper limit $\boldsymbol{a}_{\mathrm{up}}$ can be found by varying $\boldsymbol{a}$ up from $\boldsymbol{a}_{\mathrm{c}}$ until a minimum of F_{tot} at $\delta = 54.74°$ is reached. Similarly varying $\boldsymbol{a}$ down from $\boldsymbol{a}_{\mathrm{c}}$ until a minimum of F_{tot} at $\delta = 0°$ is reached the lower limit $\boldsymbol{a}_{\mathrm{low}}$ can be found. A suitable program has been written to calculate $\boldsymbol{a}_{\mathrm{c}}$ and to establish the boundaries $\boldsymbol{a}_{\mathrm{up}}(T)$ and $\boldsymbol{a}_{\mathrm{low}}(T)$ of a broad spin reorientation.

Based on the results of Sect. 2 and [24] several combinations of the values $\{K_i^{\mathrm{n}}\}$ and $\{K_i^{\mathrm{f}}\}$ have been considered. Only a limited number of the spin orientation diagrams can be presented here. Variation of the spin reorientation region for the total $Fe^{2+} - Fe^{3+}$ system with x_{e} and $\boldsymbol{h}$ is shown in Fig. 5 for a chosen combination $(\boldsymbol{n}, \boldsymbol{f})$. The corresponding results for the Fe^{2+} subsystem only, which are independent of x_{e}, are plotted for comparison in each graph with $x_{\mathrm{e}} = 0.05$. It is thus clear that any conclusion on the bulk properties of YIG : Si drawn on the basis of the Fe^{2+} subsystem considerations must be misleading.

The values δ_{mc} at the top of each graph refer to the upper critical angle (see Sect. 2.2) for the total anisotropy energy at a given temperature as indicated in Figure 5. The meaning of δ_{mc} is that it gives maximum angle that the total magnetization $\boldsymbol{M}$ makes with the direction [100] before switching sharply to the direction [111] as $\boldsymbol{a}$ approaches $\boldsymbol{a}_{\mathrm{up}}$ from below at a constant T.

The near site occupancy $\boldsymbol{a}$ should tend to values close to 1 with T tending to 0 K (see Sect. 3.1), whereas with increasing temperature a certain "saturation" value of $\boldsymbol{a}$ will be reached as determined by the crystal structure of YIG : Si and the actual content of the Fe^{2+} ions. A case when in the absence of an external disturbance, as e.g. irradiation, $\boldsymbol{a}$ reaches zero (i.e. $\boldsymbol{b} = 1$) with increasing T is rather unlikely, except for very lightly doped samples. Except for the above end criteria, nothing more specific can be said about the temperature dependence of the quantity $\boldsymbol{a}$. A possible curve $\boldsymbol{a}(T)$ which satisfies the end criteria is shown on the diagram with $x_{\mathrm{e}} = 0.30$ and $\boldsymbol{h} = 160\ \mathrm{cm^{-1}}$ (Figure 5). The importance of the spin orientation diagrams for theoretical determination of the bulk onset (T_1') and completion (T_2') temperatures is then well visualized. The values of T_1' and T_2' derived in this way are around 110 K and 250 K, respectively, both for $x_{\mathrm{e}} = 0.30$ (Fig. 5) and $x_{\mathrm{e}} = 0.25$ with $\boldsymbol{h} = 160\ \mathrm{cm^{-1}}$. Hence the predicted transition widths match quite well with the experimental ones for samples with higher Si-content (see Table 3). Figure 5 shows that the present two-center model is feasible to account for the broad spin reorientation in YIG : Si much better than the one-center model considered in Section 2. The absence of spin reorientations in samples having Si-content less than 0.18 per formula unit can also be understood in terms of our model. It implies that for low x_{e} the curve $\boldsymbol{a}(T)$ lies whole in the region "[111]", i.e. above the predicted spin reorientation boundaries and hence the magnetization would always be along the [111] direction. The experimentally observed decrease in T_1' with

decreasing x_e can be explained if one assumes different anisotropy constants $\{K_i^n\}$ and $\{K_i^f\}$ for Fe^{2+} ions in samples with different x_e.

The above spin orientation diagrams illustrate the mechanism of a spin reorientation in YIG : Si in the framework of the present two-center model. The diagrams may also be very useful for interpretation of the related phenomena in YIG : Si, as e.g. change of sign of the bulk K_1 constant [16, 36, 37] or the photomagnetic effects [18, 27, 29, 31, 32, 35]. A redistribution of Fe^{2+} ions over the near and far sites induced by irradiation [18, 29, 31] at a constant T will be reflected in the spin orientation diagrams as a variation along the ***a***-axis at a given T. The present model suggests that both abrupt and continuous spin reorientation can occur in YIG : Si due to a redistribution of Fe^{2+} ions. The nature of these transitions would depend on the temperature region.

4. Conclusions

The present study has revealed an alternative mechanism of spin reorientation in YIG : Si which is due to competition between Fe^{2+} ions on the near and far sites. Magnetic anisotropy theory based on a novel energy-level model for the Fe^{2+}-ions has been studies both in one-center and two-center approximation. The results show that the singlet model accounts for the properties of the Fe^{2+} subsystem as well as the total $Fe^{2+} - Fe^{3+}$ system considerably better than the doublet model. The singlet model with the uniform distribution of Fe^{2+} ions over the inequivalent octahedral sites exhibits both abrupt and continuous spin reorientations. Thus, this model offers an alternative explanation of the broad spin reorientation in YIG : Si with respect to the concept of temperature-independent imbalance in the site distribution necessary within the doublet model.

Our anisotropy studies show significance of the higher-order cubic anisotropy terms. It appears that these are the constants K_3 and K_4 which are responsible for the broad spin reorientations in YIG : Si. Our model accounts also for the composition dependence of the transition temperatures. The variation of the position of the minimum anisotropy energy with the angle δ and temperature indicates a peculiar feature of the overall broad spin reorientations, namely, that the transitions are "continuous" only to a certain extent. In the vicinity of the bulk completion temperature of the spin reorientation the transition is likely to be no longer "smooth" but "sharp" in nature. This suggests that more careful experiments are needed in the temperature ranges close to the completion temperatures. The spin orientation diagrams established here within the two-center model may also be useful in further studies of spin reorientation process and related phenomena in YIG : Si.

Two further developments suggest themselves. First, it would be worthwhile to consider the effects of a nonuniform distribution of Fe^{2+} ions within the framework of the singlet energy level model. Second, the consequences of a feasible coexistence of the Fe^{2+} ions in the orbital singlet and the anisotropic doublet ground state could be investigated. More studies would be necessary to check implications of these refinements, so that the superiority of either of the two possible energy level models could be established.

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Appendix

Table A1. Experimental anisotropy constants for pure YIG vs. temperature used in the present calculations in [erg/cm^3] $* 10^{-4}$ (after Hansen [9, 28]).

T(K)	$-K_1$	$-K_2$	T(K)	$-K_1$	$-K_2$
4.2	2.48	0.26	120	1.96	0.23
10	2.47	0.26	140	1.81	0.20
20	2.44	0.24	160	1.62	0.20
30	2.41	0.25	180	1.46	0.18
40	2.36	0.27	200	1.27	0.18
50	2.33	0.25	220	1.09	0.14
60	2.29	0.26	240	0.92	0.14
70	2.24	0.25	260	0.78	0.10
80	2.18	0.25	280	0.67	0.10
90	2.13	0.25	300	0.57	0.08
100	2.08	0.25	320	0.48	0.04

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