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 ($a$) of the near Fe$^{2+}$ sites. Spin orientation diagrams in the plane $a$–$T$ 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 $M$ is predicted not to rotate smoothly between the [100] and [111] direction. There exists an upper critical angle beyond which $M$ 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$_3$Fe$_{5-x}$Si$_x$O$_{12}$ [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)

$$ F_a = K_1 P + K_2 Q + K_3 P^2 + K_4 P Q + \ldots $$  \hspace{1cm} (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 ($^2A_g$) 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 below $T_c$. 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 uniaxial anisotropy also at room temperatures. To the best of our knowledge a weak uniaxial anisotropy was observed in YIG:Si only...
below 30 K [16]. Neither that aspect nor cubic anisotropy has been explicitly 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 $^5$A$_{1g}$ 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], which are, in fact, induced in YIG:Me$^{4+}$ isotropic fields [25–27] which are, in fact, induced in YIG:Me$^{4+}$ isotropic fields. Then we consider how the 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 $F_i$ of a Fe$^{2+}$ ion with the framework of our model [21, 24] depends on the eight spin Hamiltonian parameters $B(\theta, \phi)$, $B(\phi)$, $B(\theta, \phi)$, $B(\phi)$, $B(\theta, \phi)$, $B(\phi)$, $B(\theta, \phi)$, and $B(\phi)$, the molecular field $\mathbf{h} = g\mu_B\mathbf{H}$, 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. $n_i = 1$, 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 $\delta$ between the magnetization $\mathbf{M}$ and the [100] direction in the plane (011), we have

$$F_1(\delta) = F_1(\delta) - F_1(\delta = 0^\circ) \quad (2)$$

The angle $\delta$ is varied in steps $5^\circ$ between $0^\circ$ ([100]) and $50^\circ$ with the last $\delta = 54.74^\circ$ ([111]); thus $q = 1–12$ in (2). Considering a set of the $F_1(\delta)$ values, $q = 1, \ldots, 12$, for a given set of the $B(q,\phi)^2$s and $\mathbf{h}$ at a certain temperature we find a minimum anisotropy energy and the respective angle $\delta_q$. There are three different cases [24]: (a) the minimum of $|F_1(\delta_q)|$ is at $\delta_1 = 0^\circ$, i.e. $\mathbf{M}$ [100] at all $T$ between 0 and 300 K, (b) the minimum is at $\delta_{12} = 54.74^\circ$, i.e. $\mathbf{M}$ [111] at all $T$, and (c) the minimum gradually shifts from the [100] direction at low $T$ towards the [111] direction at high $T$. Table 1 illustrates the results [24] for the $B(q,\phi)^2$ sets predicted by the microscopic theory [21] and several $\mathbf{h}$ values in the range 150 to 500 cm$^{-1}$ suitable for YIG [9, 10, 11, 12].

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.
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 $(B_q^{(k)}, h)$ sets. The nontrigonal ($I$) and trigonal ($A$) crystal field parameters and the molecular field $h$ are in cm$^{-1}$.

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The case (c), i.e. a broad spin reorientation, has also been found for some other $(B_q^{(k)}, h)$ 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 $F_q^i$ for the direction [110] to be sure that it does not fall below the minimum of $F_q^i(\delta_q)$. In no case considered in Table 1 and in the subsequent calculations $F_q^i$ [110] is lower than the corresponding minimum of $F_q^i(\delta_q)$. 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 $F_q^i(\delta_q)$ and $F_q^i$ [110] with the three different values of $\lambda$ 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 $(B_q^{(k)}, h)$ 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 $B_0^{(2)} = 4.8$, $B_1^{(2)} = -2.5$, $B_2^{(2)} = -1.3$, $B_3^{(2)} = 0.0060$, $B_4^{(2)} = -0.0155$, $B_5^{(2)} = 0.0015$, $B_6^{(2)} = 0.0001$ and $B_7^{(2)} = 0.0004$ cm$^{-1}$. The anisotropy energy $F_q^i(\delta)$ is plotted vs. the angle $\delta$ 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 $h$ values.

It is seen in Fig. 2 that the set (v) with $h = 160$ and 180 cm$^{-1}$, besides a broad spin reorientation at
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 $h = 200 \text{ cm}^{-1}$ 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 $\delta_m$ at which the minimum anisotropy energy occurs at a given $T$ does not vary smoothly between $0^\circ$ and $54.74^\circ$. There exists certain upper critical angle $\delta_{mc}$ beyond which $M$ rotates abruptly within a very narrow temperature range into the direction [111] as $T$ approaches from below $T_2$. For the set (v) with $h = 160-200 \text{ cm}^{-1}$ the upper critical angle $\delta_{mc}$ is about $35^\circ$. It is seen in Fig. 1 that for $T$ close to $T_2$ the anisotropy energy $F_a(\delta)$ has no stable minimum and only a smeared minimum region between $\delta_{mc}$ and $54.74^\circ$. 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 $F_i[hhk]$ for some five chosen [hhk] 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 $(B_{ik}^{(k)}, h)$ 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. 2. Positions of the minimum anisotropy energy of Fe$^{2+}$ ion in YIG:Si vs. temperature with the $B_{ik}^{(k)}$ set (v) and various molecular fields.](image-url)

![Fig. 3. Temperature dependence of the cubic anisotropy constants for Fe$^{2+}$ ion in YIG:Si with the $B_{ik}^{(k)}$ set (v) and $h = 200 \text{ cm}^{-1}$.](image-url)
temperatures. As we focus this paper on the mechanism of spin reorientation the diagrams $K_i(T)$ for the other $(B_{ij}^{(k)}, h)$ sets will not be presented here. It is enough to say that in all cases where a broad spin reorientation is found the constants $K_1$ 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 $K_1^1$ and $K_3^1$ and $K_3^2$ [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] $K_i$ are related to the $K_i$’s [3, 10, 24] in (1) as follows [23]:

$$K_i^1 = K_1, \quad K_i^2 = K_2 + 2K_3, \quad K_i^3 = K_3.$$  \hspace{1cm} (3)

Hence, in terms of the $K_i$’s the condition [12] for the lower boundary of spin reorientation is given by $K_i^1 = 0$, $K_3 > 0$. A comparison of Figs. 2 and 3 and an inspection of the corresponding data for the other $(B_{ij}^{(k)}, h)$ sets indicates, in fact, that (i) the onset temperature $T_1$ coincides with the temperature at which $K_i^1$ changes sign and (ii) $K_3$ is always positive. Similarly the condition [12] for the upper boundary of the transition can be written as

$$L \equiv K_i^1/K_3 = -[(K_2/K_3 + 4)^2]/24 \equiv R,$$  \hspace{1cm} (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 $(B_{ij}^{(k)}, h)$ 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 $(B_{ij}^{(k)}, h)$ 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, 0.0100 for the set (ii) and (iii), respectively, whereas $B_2^{(2)} = -3.4$ for both sets. For the set (iv) with $h = 160$ and 180 cm$^{-1}$ an abrupt or very narrow spin reorientation occurs at about 250 and 286 K, respectively, whereas with $h = 200$ cm$^{-1}$ no spin reorientation is found below 320 K. The $B_{ij}^{(k)}$ parameters for the set (vi) are identical with those for the complementary set (v), except for the $B_2^{(2)}$ 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 $B_2^{(2)}$, $B_2^{(4)}$, $B_0^{(4)}$ 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.

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 \(B_{ij}^{(2)}\) shift significantly the transition region. Our model implies that in the specimens with low Si-content the Fe\(^{2+}\) ions have lower \(A\) [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.

### 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 \((f)\) and trigonal \((J)\) 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 \(B_{ij}^{(k)}\) sets (v)–(vii) as
appropriate for the near sites, while the other sets in Table 2 may be prescribed to the far sites. If the majority of Fe$^{3+}$ 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 $a$ and $b$, respectively $(a + b = 1)$. These quantities must be both temperature and composition dependant. The total anisotropy energy of the Fe$^{3+}$ – Fe$^{2+}$ system is thus given by

$$F_{\text{tot}} = F_a(\text{Fe}^{3+}) + a F_n(\text{Fe}^{2+}) + (1 - a) F_f(\text{Fe}^{2+}),$$

(5)

where the superscript $n$ and $f$ 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

$$F_{\text{tot}}(\delta) = \sum_{i=1}^{4} K_{i}^{\text{tot}} f_i(\delta),$$

(6)
where \( f_i(\delta) \) are the angle-dependent functions [23, 24] and

\[
K_i^{\text{tot}} \text{(erg/cm}^3) = K_i(\text{Fe}^{3+}) + a \bar{K}_i(\text{Fe}^{2+}) + (1 - a) \bar{K}_j(\text{Fe}^{2+}).
\]

We adopt the following conversion factor [28]:

\[
\bar{K}_i \text{(erg/cm}^3) = x_e 0.84*10^6 \bar{K}_i \text{(cm}^{-1}/\text{Fe}^{2+} \text{ion}),
\]

where \( x_e \) is the effective Fe\(^{2+} \) content per formula unit, \( p = f \) or \( n \), and \( \bar{K}_i \) 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 \( K_i(\text{Fe}^{3+}) \) 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 \( F_{\text{tot}}^{\text{of}} \) of (5) is finally a function of the following parameters: \( \delta \), \( T \), \( x_e \), \( a \), and the values \( \{K_i^f\} \equiv n \) and \( \{K_i^f\} \equiv f \) (\( i = 1, \ldots, 4 \)). Considering for a given set \( (n, f) \) \( F_{\text{tot}} \) vs \( \delta \) at a constant \( T \) and \( x_e \) the position of the minimum total anisotropy energy can be found as a function of \( a \). Then, a spin orientation diagram [11, 12] in the plane \( a - T \) can be constructed. In these calculations \( \delta \) 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 \( F_{\text{tot}}^{\text{of}} \). The variation of \( F_{\text{tot}}^{\text{of}}(\delta) \) in the vicinity of a transition is depicted in Figure 4. There exists a critical value of \( a \) for which \( F_{\text{tot}}^{\text{of}}(54.74°) \) is equal zero. With \( a = a_c \), the minimum of \( F_{\text{tot}}^{\text{of}}(\delta) \) is either at \( \delta = 0° \) and 54.74° for an abrupt transition (curve C in Fig. 4) or at an intermediate \( \delta \) for a broad transition (curve D). In the later case the upper limit \( a_{up} \) can be found by varying \( a \) up from \( a_c \) until a minimum of \( F_{\text{tot}}^{\text{of}} \) at \( \delta = 54.74° \) is reached. Similarly varying \( a \) down from \( a_c \) until a minimum of \( F_{\text{tot}}^{\text{of}} \) at \( \delta = 0° \) is reached the lower limit \( a_{low} \) can be found. A suitable program has been written to calculate \( a \), and to establish the boundaries \( a_{up}(T) \) and \( a_{low}(T) \) of a broad spin reorientation.

Based on the results of Sect. 2 and [24] several combinations of the values \( \{K_i^f\} \) and \( \{K_i^f\} \) 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 \( h \) is shown in Fig. 5 for a chosen combination \( (n, f) \). The corresponding results for the Fe\(^{2+} \) subsystem only, which are independent of \( x_e \), are plotted for comparison in each graph with \( x_e = 0.05 \). 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 \( \delta_{mc} \) 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 \( \delta_{mc} \) is that it gives maximum angle that the total magnetization \( M \) makes with the direction [100] before switching sharply to the direction [111] as \( a \) approaches \( a_{up} \) from below at a constant \( T \).

The near site occupancy \( a \) 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 \( a \) 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, \( a \) reaches zero (i.e. \( b = 1 \)) 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 \( a \). A possible curve \( a(T) \) which satisfies the end criteria is shown on the diagram with \( x_e = 0.30 \) and \( h = 160 \text{ cm}^{-1} \) (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 \( x_e = 0.30 \) (Fig. 5) and \( x_e = 0.25 \) with \( h = 160 \text{ cm}^{-1} \). 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 \( a(T) \) 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 $\{K'_i\}$ and $\{K''_i\}$ 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 studied 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 $K_4$ and $K_2$ 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 $\delta$ 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$^3$] $\times 10^{-4}$ (after Hansen [9, 28]).

<table>
<thead>
<tr>
<th>$T$(K)</th>
<th>$-K_1$</th>
<th>$-K_2$</th>
<th>$T$(K)</th>
<th>$-K_1$</th>
<th>$-K_2$</th>
</tr>
</thead>
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<tr>
<td>4.2</td>
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<td>0.26</td>
<td>120</td>
<td>1.96</td>
<td>0.23</td>
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<td>10</td>
<td>2.47</td>
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<td>140</td>
<td>1.81</td>
<td>0.20</td>
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<tr>
<td>20</td>
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<td>0.24</td>
<td>160</td>
<td>1.62</td>
<td>0.20</td>
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<td>1.46</td>
<td>0.18</td>
</tr>
<tr>
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<td>0.27</td>
<td>200</td>
<td>1.27</td>
<td>0.18</td>
</tr>
<tr>
<td>50</td>
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<td>0.25</td>
<td>220</td>
<td>1.09</td>
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</tr>
<tr>
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<td>2.29</td>
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<td>0.92</td>
<td>0.14</td>
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<td>0.25</td>
<td>260</td>
<td>0.78</td>
<td>0.10</td>
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<tr>
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</tr>
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</tr>
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<td>0.25</td>
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<td>0.48</td>
<td>0.04</td>
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