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To cite this version:

P. Hansen, W. Tolksdorf. ANISOTROPY OF RUTHENIUM-SUBSTITUTED YTTRIUM-IRON-GARNET. Journal de Physique Colloques, 1971, 32 (C1), pp.C1-200-C1-201. 10.1051/jphyscol:1971161. jpa-00214488

HAL Id: jpa-00214488
https://hal.archives-ouvertes.fr/jpa-00214488
Submitted on 1 Jan 1971

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ANISOTROPY OF RUTHENIUM-SUBSTITUTED YTTRIUM-IRON-GARNET

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Résumé. — La contribution à l'anisotropie des ions f à bas spin a été calculée avec le modèle à un ion. Une comparaison de la théorie avec des mesures d'anisotropie de YFeO$_3$(YIG) dopé avec Ru$^{3+}$(4d$^5$) montre qu'on peut obtenir un bon accord dans le régime de température de 4.2 °K à 300 °K.

Abstract. — The anisotropy contribution of low spin f$^5$-ions has been calculated by means of the simple-ion model. The theoretical expression can be well fitted to anisotropy measurements of Ru$^{3+}$(4d$^5$)-doped Y$_2$Fe$_5$O$_{12}$(YIG) in the temperature range 4.2 °K to 300 °K.

In order to investigate the influence of ruthenium on the anisotropy of yttrium iron garnet, single crystals of the composition Y$_{3-x}$Ca$_x$Fe$_{5-x}$Ru$_x$O$_{12}$ [1] with various values of $x$ and $y$ were grown from a PbO/PbF$_2$/BaO$_3$ flux. A method was used which separated the crystals from the flux under growing conditions between 1 100 °C and 1 050 °C [2]. The concentration of strong relaxing cations such as Co$^{3+}$, Tb$^{3+}$, Dy$^{3+}$ etc. in the starting materials was certainly lower than 1 ppm. A typical composition of the melt was e. g. 39.02 g PbO; 32.89 g PbF$_2$; 1.81 g BaO; 0.03 g CaO; 2.61 g Fe$_2$O$_3$ and 24.64 g Y$_{2.97}$Ca$_{0.03}$Ru$_{0.01}$Fe$_{1.99}$O$_{12}$ from which we obtained crystals with $x < 0.01$ and $y = 0.01$.

The contribution of a substituted ion to the free energy per unit volume is given by [3]

$$ F = -kT \sum_i N_i \ln z_i, \quad (1) $$

where

$$ z_i = \sum_j \exp \left( -\frac{E_i}{kT} \right). \quad (2) $$

The sum in Eq. (1) runs over all magnetically inequivalent sites and in Eq. (2) over the various energy levels of the ion at site $i$, and $N_i$ represents the population of this site. The anisotropy contributions are related to the principal directions of the free energy (1) according to

$$ \Delta K_1 = 4(F(110) - F(100)) $$

$$ \Delta K_2 = 9(3F(111) + F(100) - 4F(110)) \quad (3) $$

if the higher order terms in Eq. (1) can be neglected.

The cubic ground state of Ru$^{3+}$(4d$^5$) at octahedral sites in the garnets is the low spin state $^2T_{2g}$ with the total spin $S = \frac{1}{2}$ [1], [4]. This sixfold degenerate term is split up by the local trigonal field, the spin-orbit coupling, and the exchange field, as shown in figure 1. The corresponding Hamiltonian is given by

$$ \mathcal{H} = V(ns) + jL.S + g_H S.H. \quad (4) $$

For a low spin d$^5$-configuration $\lambda$ can be expressed by the one-electron spin-orbit coupling parameter $\xi$ as $\lambda = -\xi$ because this configuration can be regarded as a hole in the $t_{2g}$-shell. The exchange interaction is considered in the limits of the molecular field model and is assumed to be isotropic. From the representations of the trigonal double group it follows that the degeneracy of $^2T_{2g}$ is removed and split up into three magnetic doublets. The lowest level is well separated in a wide range of $\nu/\zeta$ values ($\nu$ is the one-electron trigonal field parameter). It is therefore sufficient to study the splitting of this level due to the exchange field $H_e$. The wave functions of this doublet are given by

$$ \psi_+ = a | 0, \frac{1}{2} > + b(\sqrt{\frac{3}{2}} | 2, -\frac{1}{2} > + \sqrt{\frac{1}{2}} | 1, -\frac{1}{2} >) \quad (5) $$

$$ \psi_- = -a | 0, -\frac{1}{2} > + b(\sqrt{\frac{3}{2}} | 2, \frac{1}{2} > - \sqrt{\frac{1}{2}} | 1, \frac{1}{2} >), $$

where

$$ a^2 + b^2 = 1, a^2 = \frac{1}{2} \left( 1 - \frac{\nu/\xi}{\sqrt{2 + (\frac{1}{2} - \nu/\xi)^2}} \right). $$

The numbers in the ket are the $m_L$ and $m_S$ values.
of the \( d \) functions and the spin functions, respectively. The axis of quantization is the local trigonal axis which coincides with one of the [111]-directions.

Applying the Hamiltonian to Eq. (5) leads to the two energies:

\[
E_\pm(\gamma_i) = E(v, \xi) \pm \frac{1}{2} \Delta E(\gamma_i),
\]

(6)

where

\[
\Delta E(\gamma_i) = g\mu_B H_e \sqrt{1 - 2 a^2 \cos^2 \gamma_i + a^2 \sin^2 \gamma_i}.
\]

\( \gamma_i \) represents the angle between the magnetization \( \mathbf{M} \) and one of the four possible [111]-directions. \( E(v, \xi) \) does not depend on the direction of \( \mathbf{M} \) and thus does not contribute to the anisotropy. \( \Delta E(\gamma_i) \) are given in figure 2. From Eqs. (1), (2), (3), and (6) the anisotropy can be calculated.

![Exchange splitting of the lowest magnetic doublet of low spin \( d^5 \) ions.](image)

Anisotropy measurements of Ru-doped YIG of the composition \( \text{Y}_{3-x}\text{Ca}_{x}\text{Fe}_{5-x}\text{Ru}_x\text{O}_{12} \) [1] have been performed by the method of ferromagnetic resonance at 9.25 GHz and in the temperature range of 4.2 °K to 300 °K. For \( x \ll 1 \) the influence of \( x \) on the saturation magnetization can be neglected and \( M_s(T) \) of the pure YIG can be used. The results are given in figure 3. On account of the calcium content, Ru\(^{4+} \) and Fe\(^{3+} \) may be assumed to be present in this material besides Ru\(^{3+} \) [5]. The anisotropy contribution of Fe\(^{4+} \), however, is negligible [6], and the concentration of Ru\(^{4+} \) is very small [1]. Therefore, the anisotropy contributions can be explained in terms of Ru\(^{3+} \) alone, and the given theory can be applied. A good fit in the whole temperature range is obtained for \( v/\xi = -0.95 \) and \( gH_e = 1.2 \times 10^7 \) Oe. The value of \( v/\xi \) agrees well with \( v/\xi = -1.06 \) obtained from ESR measurements of YGaG [4]. The exchange interaction Ru\(^{3+} \) – Fe\(^{3+} \) is slightly higher than that of the iron ions. If an anisotropic \( g \)-factor is used in Eq. (4), the \( v/\xi \) value is reduced to \( v/\xi = -0.6 \) [1].

\[\text{Fig. 3. — The contribution } \Delta K_i = K_i(x) - K_i(0) \text{ of Ru}^{3+} \text{ to the anisotropy of } \text{Y}_{3.45}\text{Ca}_{0.01}\text{Fe}_{5-x}\text{Ru}_x\text{O}_{12} \text{ for } x \approx 0.005.\]

![Graph](image)

References