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Umbrella Structure in Pr, Nd and Yb Substituted YIG Studied by ^{57}Fe NMR

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Abstract. The NMR spectra of ^{57}Fe in Pr:YIG, Nd:YIG and Yb:YIG were measured on polycrystalline samples at 4.2 K in the zero external field. In the NMR spectra of both tetrahedral and octahedral sites the resolved satellite structure appears. This structure is caused by the change of the hyperfine field on the nuclei of those Fe^{3+} ions, which are the nearest or next nearest neighbours of the substituted Re^{3+} ion. The change of the hyperfine field is given by the change of dipole and transferred contribution. Both contributions are sensitive to the kind of Re^{3+} ion and to the value and direction of its magnetic moment. The satellite structure of NMR spectra provides sufficient amount of information (splitting) to find unknown parameters of the non-collinear (umbrella) arrangement of Re^{3+} magnetic moments.

Yttrium iron garnet $(\text{Y}_3)_c\{\text{Fe}_2\}_d\{\text{Fe}_3\}_a\text{O}_{12}$ (YIG) is one of the often studied ferrimagnetic cubic oxides. There are three types of cation lattice sites in YIG. Fe^{3+} ions occupy trigonal distorted octahedral a-sites with the symmetry axis along the [111] direction and tetragonally distorted tetrahedral d-sites having the symmetry axis along the [100] direction. The local symmetry of dodecahedral c-sites occupied by Y^{3+} ions is orthorhombic and there are six c-sites in the unit cell with the different orientation of the local co-ordinate system.

The principal magnetic interaction in YIG is the superexchange in the $(\text{Fe}^{3+})_d\text{-O}^{2-}\text{-(Fe}^{3+})_a$ triads. This interaction forms two antiparallel iron sublattices with the easy axis along the [111] direction. In rare earth substituted systems $(\text{Y}_{3-x}\text{Re}_x)_c\{\text{Fe}_2\}_d\{\text{Fe}_3\}_a\text{O}_{12}$ (Re:YIG) reported in this paper the easy axis keeps the [111] direction of YIG.

Magnetic moments of Re-cations in Re:YIG have at most six different magnitudes and directions if the magnetization lies in the general direction. If the magnetization lies in the [111] direction (in zero external field) there exist two groups of three magnetically equivalent c sites (c, c') [1]. Re-magnetic moments lie in the planes of the (110) type, forming so called umbrella structure [1] described by magnitudes (m, m') of Re-moments and umbrella angles (φ , φ'). Umbrella angles are measured from [111]. In our notation the angle is positive when turning from [111] to [112] in the (110) plane and similarly in other planes of the (110) type.

The ^{57}Fe NMR spectrum in the stoichiometric YIG consists of three lines corresponding to Fe^{3+} ions in d-sites and two magnetically inequivalent a₁- and a₂-sites [2]. When a small amount of Y^{3+} ions is replaced by Re^{3+} ions, characteristic satellite structure in the NMR spectrum appears. This structure is caused by the change of the hyperfine field on the nuclei of Fe^{3+} ions which have the Re^{3+} ion in the first or second neighbourhood. Re-ions in the third and other environments contribute to the line broadening only (in our region of Re-concentration). The value of the splitting is sensitive to the ionic radius of the Re^{3+} ion, to the magnitude and orientation of its magnetic moment and to its position in the vicinity of the given Fe^{3+} ion.

In this paper we present the values of Re-magnetic moments and umbrella angles obtained from the analysis of ^{57}Fe NMR spectra in Pr:YIG (x=0.1), Nd:YIG (x=0.35) and Yb:YIG (x=0.1).

Our NMR experiments were performed on the polycrystalline samples prepared by the conventional ceramic technology and sintered at about 1450 °C. The phase purity and homogeneity was checked by X-rays. Concentration of Re was given by the technology and checked by the atomic spectroscopy and by the amplitude of satellites. The estimated error in x was of the order of 0.01.

NMR spectra were measured by the spin echo technique at 4.2 K. The phase coherent spectrometer with averaging technique and complex Fourier transformation was used. As the NMR line splitting is small compared with the frequency of the main lines, no frequency corrections were made. Relative accuracy of the amplitude in the NMR spectrum is then better than 5%. All spectra were measured with sufficiently high rf-power to excite the domain signal only. Examples of the NMR spectra normalised to 100 are shown in the Fig. 1.

To fit the satellite structure, we have supposed the independent and additive contribution of each Re-defect, the random distribution of defects in the c- and c'-sites and the same shape of the main and satellite lines.

The splitting between the main line and satellites (Δv_{exp}) at low temperatures can be written as a sum of three contributions [3]: $\Delta v_{\text{exp}} = \Delta v_e + \Delta v_{\text{pol}} + \Delta v_{\text{dip}}$. The change of the crystal environment contribution (Δv_e), independent of

Figure 1: NMR spectra of ^{57}Fe in Re:YIG, at 4.2K, $B_{\text{ex}}=0$. a) d-sites, b) a_1 - and a_2 - sites.

the Re-magnetic moment, was determined using the linear dependence of Δv_c on the ion radius of Re^{3+} in the eight-fold co-ordination [3], [4]. To determine the polarised part of the crystal environment contribution (Δv_{pol}), it was supposed, that this contribution is proportional to the projection of the total Re^{3+} -spin on the direction of the Re^{3+} -magnetic moment. Using the value of $\Delta v_{\text{pol}}(\text{Gd})$, determined from the temperature dependence of the NMR spectra in Gd:YIG [5], we can calculate: $\Delta v_{\text{pol}}(\text{Re}) = -\Delta v_{\text{pol}}(\text{Gd}) \cdot m \cdot \cos\phi \cdot (g_i - 1) / 3.5 g_i$ ([3], [4]). The dipolar contribution (Δv_{dip}) is given by the projection of the dipolar field on the direction of the ^{57}Fe hyperfine field and can be calculated Δv_{dip} using the lattice structure data and two pairs of unknown umbrella parameters (m, ϕ) and (m', ϕ').

To find these parameters we need at least two pairs of independent splitting in the NMR spectrum. In our case we have used two well resolved satellites of the d-line (s_{d1}, s_{d1}') and the satellites of the a_1 - and a_2 -lines (s_{a1}, s_{a2}') which correspond to the substitution of the Re^{3+} in the nearest c (c') - sites of the given d- or a-site respectively. The results are summarised in the Table 1. Since we can not determine with confidence which of the d-line satellites belongs to the c- or c' - site, we have calculated two sets of umbrella parameters. To distinguish between these two sets the higher resolution in

Tab.1.

Re	$m(\mu_B)$	$\phi(^{\circ})$	$m'(\mu_B)$	$\phi'(^{\circ})$	$m[111]_{\text{NMR}}$	$m[111]_{\text{mag}}$ [Ref.]
Pr	2.5 (2)	9 (3)	3.4 (3)	-36 (5)	2.6 (3)	1.53 [8]
	3.1 (3)	-40 (8)	2.7 (3)	8 (3)	2.6 (3)	
Nd	1.8 (2)	19 (5)	0.9 (2)	-12 (3)	1.3 (2)	1.65 [8]
	1.7 (2)	-13 (4)	1.2 (2)	40 (5)	1.3 (2)	
Yb	2.0 (2)	217 (5)	1.7 (2)	232 (5)	1.3 (3)	1.67 [8]
	2.0 (2)	220 (5)	1.6 (2)	227 (5)	1.3 (3)	
	1.7 *	186 *	1.7 *	187 *		

the NMR spectra (lower Re concentration) is necessary to see two other satellites of the a_2 -line [2], or the second nearest neighbour satellite of the a_1 -line [3].

The mean magnetic moments along the [111] direction deduced from the NMR data ($m[111]_{\text{NMR}}$) for Nd^{3+} and Yb^{3+} in YIG compare favourably with the values deduced from the saturation magnetization ($m[111]_{\text{mag}}$) [6] and infrared spectroscopy [7]. A similar comparison of Pr^{3+} results shows a considerable discrepancy, which can be tentatively associated with a special nature of the crystal-field (CF) ground state [8]. The quantitative calculations involving the CF potential and the magnetic exchange interaction Pr-Fe are in progress. These should allow us to decide which of two above mentioned umbrella structures compatible with our NMR data in Pr:YIG is correct.

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