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MAGNETIZATION ANOMALIES IN Ca²⁺(Fe⁴⁺) DOPED YIG DILUTED WITH Ga OR Sc

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Abstract. – The measurements of the temperature dependence of the magnetization of Ca²⁺M³⁺ : YIG (M³⁺ = Ga, Sc; 0.1 ≤ x ≤ 0.3; 0 ≤ y ≤ 1.5) single crystal films has shown a 5-100 % decrease of the magnetization at low temperatures compared to the two-sublattice Néel-model. The anomalies are due to charge compensating Fe⁴⁺ ions, formed via a temperature dependent localization process of the extra hole introduced by the Ca²⁺; the canting of the unsubstituted sublattice; and the low temperature ordering of the paramagnetic Fe⁴⁺ ions having less than two magnetic neighbors.

1. Introduction

The yttrium iron garnet \( \{Y^{3+}\}_{3} \{Fe^{3+}\}_{3}O_{12} \) (YIG), having antiferromagnetically coupled octahedral [Fe] and tetrahedral (Fe) sublattices, is a classical example of ferrimagnetism. Dilution with non-magnetic cations of the tetrahedral and octahedral sublattices results in the change of the magnetization and the Curie temperature. No change in the magnetic properties is expected for non-magnetic dilution at the non-magnetic yttrium sites. However, instead of the expected simple dilution effects a low temperature compensation point of the magnetization was observed in Ca²⁺Ge⁴⁺-substituted epitaxial YIG films, having Ca²⁺ > Ge⁴⁺ [1]. The electrical conductivity, the magnetic anisotropy energy and the linewidth of the ferromagnetic resonance were reported to be anomalously high [2, 3]. These effects are due to Fe⁴⁺ ions, charge compensating for the excess Ca²⁺ [1]. In this model the extra hole is delocalized at high temperatures and it becomes localized at tetrahedral Fe³⁺ sites at low temperatures, producing Fe⁴⁺ ions according to the equation:

\[
[Fe^{4+}]_T = [Fe^{4+}]_0 [1 - \exp (-T_0 / T)],
\]

where \([Fe^{4+}]_0\) is the concentration of Fe⁴⁺ at temperature \(T\), and \(T_0\) is the temperature characterizing the localization of the hole.

The goal of this study was to investigate the magnetic effects of Fe⁴⁺ in the simple Ca²⁺ : YIG system, in Ca²⁺Ga³⁺ : YIG with tetrahedral dilution and in the octahedrally diluted Ca²⁺Sc³⁺ : YIG systems.

2. Experiments

The temperature and angular dependence of the magnetization of epitaxial garnet films, grown on (111) oriented non-magnetic Gd₃Ga₅O₁₂ substrates, was investigated between 4.2 K and the Curie temperature in a vibrating sample magnetometer (PAR 155), in magnetic fields up to 13 kG. The composition of the samples has been determined by Electron Probe Microanalysis (EPMA), with an accuracy of ± 3 %. The EPMA results indicated that in the Ca-substituted samples the smaller dodecahedral Y and Lu ions occupy octahedral sites. Data for representative samples are given in table I.

Table I. – Composition, magnetization \(4\pi M_0\) at \(T = 0\) K and Curie temperature \(T_c\) of substituted garnet films.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Ca</th>
<th>Ga</th>
<th>Sc</th>
<th>Lu</th>
<th>(4\pi M_0)</th>
<th>G</th>
<th>(T_c)</th>
<th>K</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. YIG</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>2 450</td>
<td>549</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2. N10</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>1 600</td>
<td>525</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3. HP13</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>4 00</td>
<td>419</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4. BP12</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>1 706</td>
<td>243</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5. WP13</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>1 85</td>
<td>185</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

3. Results and discussion

The temperature dependence of the magnetization of a Ca: YIG and a CaGa:YIG film is shown in figure 1. For comparison, the magnetization of YIG, (expected for an \(Y_3-xCa_xFe_5O_{12}\) sample), is also shown (No. 1-3 of Tab. I). A decrease of the magnetization at low temperatures is observed due to the presence of charge compensating Fe⁴⁺ ions. A computer fit to the model of reference [1] gives very good agreement between the measured and calculated magnetization values for sample 2 with \(Fe^{4+}/Ca^{2+} = 0.4\) and \(T_0 = 25\) K. The remaining charge is compensated by oxygen vacancies and \(O^-\) ions [4]. The temperature dependence of the

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magnetization of $Y_{3-y}Ca_yFe_{5-y}Ga_yO_{12}$ (CaGa: YIG) films with $y < 1$ can be fit to the same model with similar parameters. At higher dilutions, e.g. sample number 3, the fit is good at intermediate temperatures ($Fe^{3+}/Ca^{2+} = 0.2$, $T_0 = 50$ K), however at $T_k = 35$ K the magnetization drops to zero.

Low temperature magnetic anomalies, similar to the case of Ga substituted samples, were observed in the temperature dependence of the magnetization of CaSc: YIG samples with $0 \leq Ca \leq 0.3$ and $0.6 \leq Sc \leq 1.2$ [5]. Typical examples are shown in figure 2 (No. 4 and 5). Sample 4 does not contain Ca at all, but its magnetization is decreased by about 800 G at 0 K.

For number 5 a zero magnetization was observed at $T < 10$ K. In this case, cooling in a 10 kG field through the Curie point resulted in an induced magnetization of 550 G at $T < 4.2$ K. These CaSc: YIG samples contain a high amount of the octahedral substitution (see Tab. I), and for Sc > 0.7 a localized random canting takes place [6]. Based on the theoretical models of canting, the observed decrease of the magnetization on lowering the temperature is unexpected. However, in the case of a high dilution a large number of the $Fe^{3+}$ ions, having less than two magnetic neighbors, become isolated magnetically and will behave as paramagnetic ions at room temperature. These $Fe^{3+}$ ions will order at low temperatures in the exchange field of the magnetic sublattices, contributing to the observed magnetic moment. The presence of Ca$^{2+}$ ions leads to a magnetization contribution from $Fe^{4+}$ ions, as in the case of CaGa: YIG.

In conclusion, the measured magnetization of highly diluted CaSc: YIG is a superposition of the canted ferrimagnetic moment of the Sc: YIG, the low temperature contributions from the ordered paramagnetic $Fe^{3+}$ ions and the $Fe^{4+}$ sublattice. The limit for canting for tetrahedral substitution is much higher than for octahedral substitution ($Ga^{3+} > 1.5$), so canting is not expected for the measured samples, but for number 3 the effect of the paramagnetic $Fe^{3+}$ ions can't be ignored. The measured $M_s = 0$ at $T < 0$ K for highly diluted Ga and Sc-substituted samples (No. 3 and 5) might be due to $Fe^{4+}$ ions leading to the dominance of the intrasublattice antiferromagnetism at low temperatures. The presence of $Fe^{4+}$ with high single ion anisotropy indicates a tendency toward canting for tetrahedrally diluted systems, the opposite being true for octahedral dilution.

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References