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Exchange constants in ferrimagnetic garnets

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Abstract. The exchange constants in the ferrimagnetic garnets $R_3Fe_5O_{12}$ ($R=Y^{3+}$, Gd^{3+} and Dy^{3+}) have been obtained from the experimental data on saturation magnetisation and inverse susceptibility. The sign and magnitude of the exchange constants in $Y_3Fe_5O_{12}$ have been explained on the basis of Anderson's theory of superexchange. The low temperature magnetisation data in $Dy_3Fe_5O_{12}$ have been explained by assuming canting on the c sublattice.

Keywords. Ferrimagnetic garnets; exchange constants; molecular field constants; transfer integrals; superexchange theory.

1. Introduction

The exchange constants in ferrimagnetic garnets have been obtained by many workers (Aléonard 1960; Dionne 1970; Dionne 1976; Anderson 1964) from either the magnetisation (M_s) or the inverse susceptibility (χ^{-1}) data using the molecular field approximation, but the same set of exchange constants has not been used to fit both. We have attempted to fit both the M_s and χ^{-1} data using a single set of exchange constants for the garnet systems, $Y_3Fe_5O_{12}$, $Gd_3Fe_5O_{12}$ and $Dy_3Fe_5O_{12}$, using the molecular field approximation. This is important since it is found that the M_s vs T curves can be fitted with more than one set of exchange constants. This is also true in the case of the χ^{-1} vs T curves. But of these only one set fits both the M_s and χ^{-1} data and this set has been taken to be the true set of exchange constants. The sign and magnitude of the exchange constants have been explained on the basis of the Anderson's theory of super exchange (Anderson 1959). The data on YIG and GdIG could be fitted assuming collinear spin arrangement but in the case of DyIG the saturation magnetisation at 0 K calculated from the Néel model is different from that observed experimentally. This discrepancy has been attributed by Dionne (1976) to canting within the c-sub-lattice, which is assumed to arise from the strong anisotropy field of the Dy^{3+} ions as compared to the exchange field on the c-sub-lattice. However, his assumption of constant canting from 0 K to T_c is not justified since both the exchange and anisotropy fields are temperature-dependent. We have shown that the canting angle falls rapidly with temperature and at about 30 K it vanishes and the system becomes Néel type.

2. Magnetic ordering and exchange constants

We have used the method of calculating the exchange constants from the data on M_s and χ^{-1} in systems with three collinear sub-lattices discussed elsewhere (Srivastava *et al* 1979). The substitution of magnetic rare earth ions in place of Y^{3+} in YIG is not expected to affect the exchange interactions between the transition metal ions in YIG. We have therefore used the same values of the J_{aa}, J_{ad} and J_{dd} for fitting the experimental data. The values of J_{ac}, J_{dc} and J_{cc} are one order of magnitude smaller but the susceptibility data is a sensitive function of these parameters.

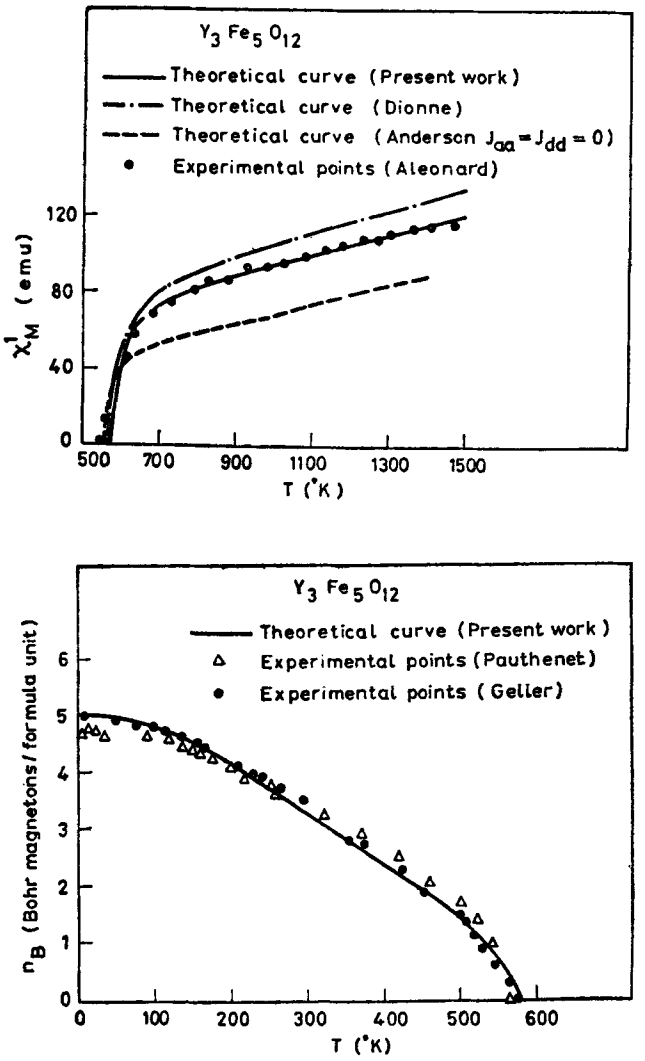

Figure 1. Theoretical and experimental curves for (a) saturation magnetisation (M_s) and (b) inverse susceptibility as a function of temperature for YIG. The theoretical curve has been calculated using the exchange constants listed in table 1.

The theoretical curves for M_s and χ^{-1} for YIG and GdIG are given in figures 1 and 2. The experimental points are from Aléonard (1960), Geller *et al* (1963, 1965) and Pauthenet (1957). The spin arrangement throughout the temperature range is Néel type.

In the case of DyIG the curve obtained on the collinear spin arrangement agrees with the experimental data only above 30 K. Below this temperature the spin arrangement is canted, the canting angle varying from 45° at 0 K to 0° at 30 K. This contrasts with the results of Dionne (1976) who assumed a temperature-independent canting of the rare earth spins, due to the strong anisotropy field. The anisotropy constant K_1, however is strongly temperature-dependent (Pearson 1962) and hence the canting angle is not likely to be temperature-independent. The observed variation of the canting angle with temperature is given in figure 3.

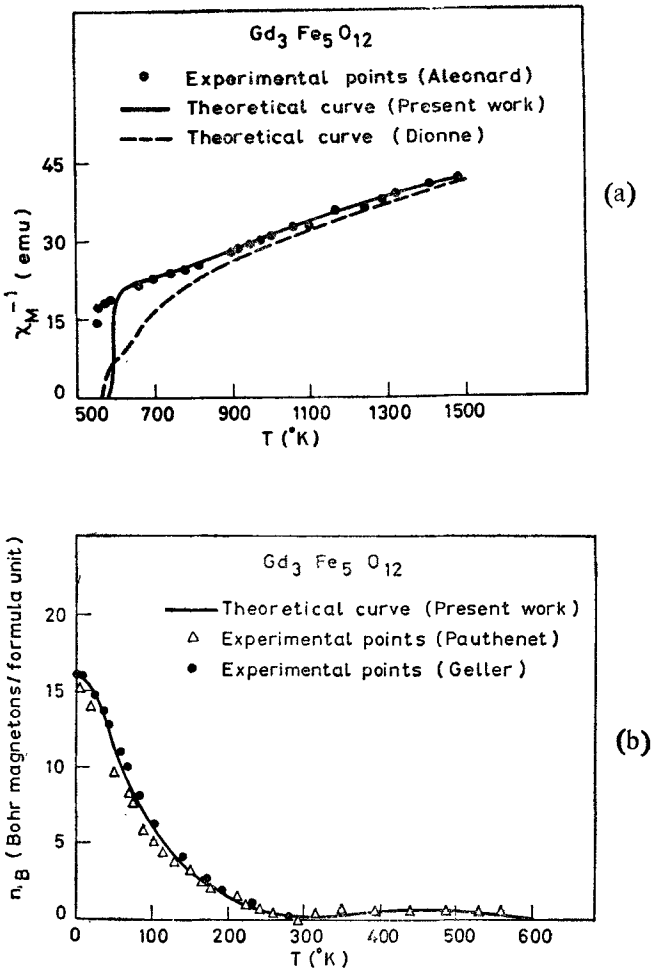

Figure 2. Theoretical and experimental curves for (a) saturation magnetisation (M_s) and (b) inverse susceptibility as a function of temperature for GdIG. The theoretical curve has been calculated using the exchange constants listed in table 1.

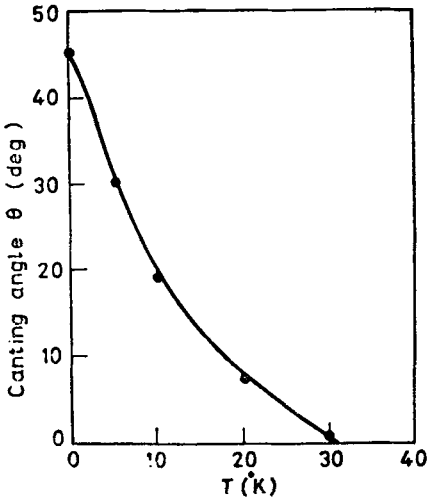

Figure 3. Variation of canting angle with temperature for Dy IG.

The fitting of the M_s and χ^{-1} data for DyIG with temperature-dependent canting angle is given in figure 4.

6. Results and discussion

The exchange constants for the three garnet systems listed in table 1 show that the magnitudes of the transition ion interactions, J_{aa}, J_{ad} and J_{dd} are greater than the transition ion-rare earth ion interaction as well as the rare earth-rare earth ion interaction. This is as expected on the basis of the overlap of the magnetic electrons on the wave functions of the ligands. On comparing the bond angles and distances of the exchange interactions in $Y_3Fe_5O_{12}$ and Fe_3O_4 it is found that the 180° *a–d* interaction in YIG is equivalent to the 180° *A–B* interaction in Fe_3O_4 and the 90° *d–d* interaction in YIG is equivalent to the 90° *A–A* interaction in Fe_3O_4. Thus it is possible that the transfer integrals obtained for Fe_3O_4 by Srivastava *et al* (1979) can be used for YIG for the 180° d^5–d^5 interaction,

$$J_{ad} = -\frac{1}{25}\left(\frac{2b^2_{\sigma\sigma}}{U} + \frac{4b^2_{\pi\sigma}}{U}\right). \tag{1}$$

The values of $b_{\sigma\sigma}$, $b_{\pi\pi}$ and U for the spinels are 0·31 eV, 0·18 eV and 10 eV respectively. This leads to a value of -28 K for spinels which closely agrees with the value of $-30{\cdot}4$ K for the garnet systems given in table 1. Similarly for the 90° d^5–d^5 interaction the value for the spinel ferrites is -14 K, which once again is close to the value of $-12{\cdot}05$ K obtained for garnets.

The ground state of Dy^{3+} has been studied by Grünberg *et al* (1969). There are a number of low lying excited states which can be occupied as the temperature is raised above 0 K. The low temperature (4·2 K) paramagnetic spectrum of Dy^{3+} in YAG and LAG observed by Ball *et al* (1963) shows a highly anisotropic

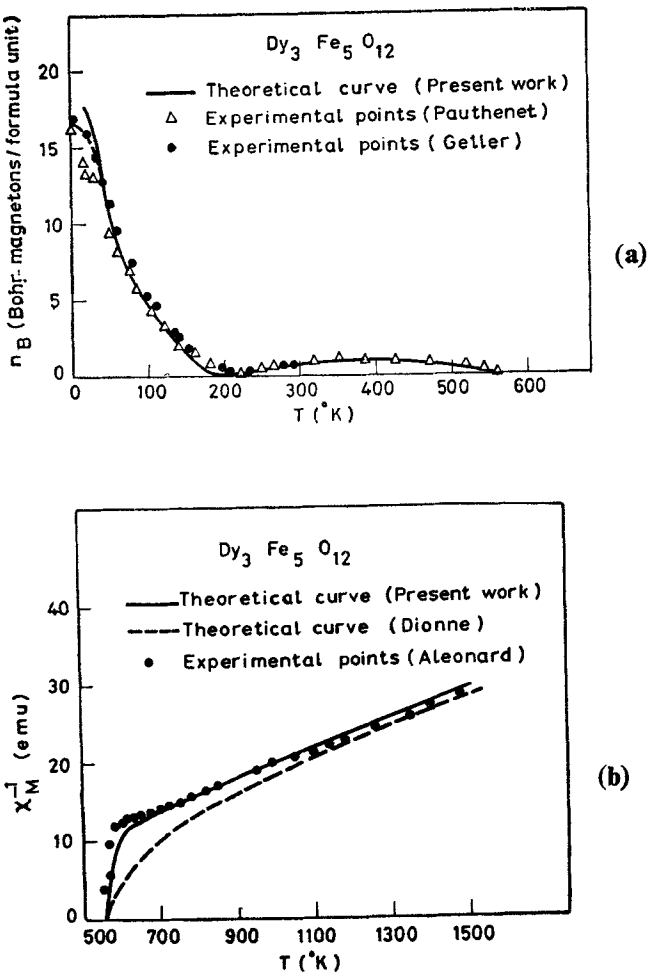

Figure 4. Theoretical and experimental curves for (a) saturation magnetisation (M_s) and (b) inverse susceptibility as a function of temperature for Dy IG. The theoretical curve has been calculated using the exchange constants listed in table 1 with canting in the *c*-sublattice, the canting angle varying with temperature as shown in figure 3.

Table 1. Exchange constants in yttrium, gadolinium and dysprosium garnets (K).

	J_{aa}	$J_{a\bar{d}}$	$J_{\bar{d}\bar{d}}$	J_{ac}	$J_{\bar{d}c}$	J_{cc}
$Y_3Fe_5O_{12}$	−6·45	−30·4	−12·05	0	0	0
$Gd_3Fe_5O_{12}$	−6·45	−30·4	−12·05	−0·6	−1·8	0
$Dy_3Fe_5O_{12}$	−6·45	−30·4	−12·05	−0·99	−0·99	0·10

behaviour. This indicates that the single ion anisotropy is large at low temperature, but as the temperature is raised on account of the occupancy of the low lying excited states, the anisotropy decreases sharply. This is supported by the observed behaviour of K_1 with temperature for YIG (Iida 1967) and DyIG (Pearson 1962). At $T = 0$, the values of K_1 for YIG and DyIG are -25×10^3 and $-2{\cdot}2 \times 10^7$ ergs/cm^3, while at $T = 77$ K these values are $-22{\cdot}4 \times 10^3$ and -970×10^3 ergs/cm^3, indicating that the single ion anisotropy of Dy^{3+} is highly temperature sensitive. The observed temperature dependence of the canting angle can therefore be interpreted to arise from the extreme anisotropy of the Dy^{3+} ion.

4. Conclusions

The exchange constants in YIG, GdIG and DyIG have been obtained from the magnetic data and interpreted in terms of the superexchange theory of Anderson.

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