Dzyaloshinskii-Moriya interaction constant in iron-gallium borate single crystals

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Abstract. The Dzyaloshinskii-Moriya interaction constant for iron-gallium borate FeₓGa₁₋ₓBO₃ single crystals has been calculated on the basis of considering antisymmetric exchange in a pair of nearest-neighbouring magnetic ions. Both numerical and analytical calculations predict a square-law dependence of the Dzyaloshinskii-Moriya constant on the contents of magnetic ions, x, in the crystals.

1. Introduction

Recently, in the Crystal Growth Laboratory at the Crimean Federal University, high quality single crystals of iron-gallium borates, FeₓGa₁₋ₓBO₃, with 0 ≤ x ≤ 1, have been synthesised [1]. All these crystals are isostructural to iron borate [1]; meanwhile, their physical properties are very different, depending on the contents of magnetic ions, x.

Iron borate, FeBO₃, x = 1, is an easy-plane weakly ferromagnetic antiferromagnet, with the Néel temperature of 348 K [2]. It possesses a number of outstanding physical characteristics and unusual its combination, for instance, it is magnetically-ordered, albeit possessing a transparency window in the visible spectral range [2]. From the viewpoint of applied research, FeBO₃ is an extremely “high-tech” material with a wide possibilities of practical applications, in particular, in the field of modern synchrotron technologies [3].

The crystals with x < 1 are, in fact, diamagnetically diluted iron borates; they all remain transparent for visible light; meanwhile, their magnetic properties depend on the degree of dilution. The possibility of modifying iron contents greatly increases their interest for fundamental research in magnetism; indeed, it allows to elucidate detailed mechanisms of transformation of the magnetic structure under the transition from magnetically ordered state to non-magnetic one [4-7]. On the other hand, the use of mixed iron-gallium borate crystals, instead of pure iron borate, is expected to significantly optimize the characteristics of technical devices, e.g., in the field of synchrotron technologies [8].

Obviously, in this context, theoretical and experimental studies of fine magnetic characteristics of iron-gallium borates become of paramount importance. We have already reported the results of electron magnetic resonance (EMR) [4], nuclear magnetic resonance [6] and theoretical studies [7, 9] of these crystals; and the aim of the present work is to carry out numerical calculations of the Dzyaloshinskii-Moriya interaction constant for FeₓGa₁₋ₓBO₃ crystals. This interaction determines the weak ferromagnetism of these crystals and of some other trigonal antiferromagnetics [10, 11].
2. Crystal and magnetic structures of iron-gallium borate crystals

The crystal structure of FeBO$_3$ is described by the space symmetry group D$_3d$. The corresponding rhombohedral unit cell is shown in Figure 1(a) [2]. Iron borate possesses two magnetic sublattices; the magnetic moments of ferric ions Fe$_1^{3+}$ and Fe$_2^{3+}$ belonging to these sublattices, $\mu_1$ and $\mu_2$, see Figure 1(a) are almost antiparallel and lie in the basal plane of the crystal perpendicular to the threefold axis C$_3$ [2]. The sublattice magnetizations are defined as:

$$M_{i,2} = \frac{1}{2} n \mu_{i,2}$$

where $n = 2.236 \times 10^{28}$ m$^{-3}$ is the iron concentration, $|M_1| = |M_2| = M_0 = 520$ G at 0 K [2].

A slight break of antiparallelism between $\mu_1$ and $\mu_2$ (the tilt angle ca. 55° [12]) produces a weak ferromagnetic moment $M = M_1 + M_2$, apart from a strong antiferromagnetic moment $L = M_1 - M_2$, see Figure 1(b). In further calculations, we use the reduced ferromagnetic $m$ and antiferromagnetic $l$ vectors:

$$m = \frac{1}{2} \frac{M}{M_0} \quad \text{and} \quad l = \frac{1}{2} \frac{L}{M_0}$$

Note that $m \perp l$, $m \ll l$, $m^2 + l^2 = 1$ and both $m$ and $l$ lie in the basal plane of the crystal.

![Figure 1](image1.png)

**Figure 1.** Crystal and magnetic structure of iron- and iron-gallium borates.

In pure iron borate, the nearest environment of an iron ion consists of $Z = 6$ irons located at a distance of 3,601 Å [2] in the planes above and below the given ion. In diamagnetically diluted Fe$_x$Ga$_{1-x}$BO$_3$ single crystals, a part of iron ions is randomly substituted by diamagnetic gallium ions; therefore, the number of nearest magnetic neighbours of Fe$^{3+}$ ion decreases, see Figure 1(c).
3. Dzyaloshinskii-Moriya constant in diamagnetically diluted crystals

Considering only the nearest magnetic neighbours, the density of the Dzyaloshinskii-Moriya energy for diamagnetically diluted crystals can be expressed as follows:

\[ E_{\text{DM}}^{\text{mix}} = \frac{1}{2} \frac{d_{\text{ex}}}{V} \sum_{i=1}^{N} Z_i \cdot (\mathbf{\mu}_i, \mathbf{\mu}_j, \mathbf{u}) \]  

(3)

where \((\mathbf{\mu}_i, \mathbf{\mu}_j, \mathbf{u})\) is the scalar triple product of the magnetic moments of iron ions belonging to the two sublattices and \(\mathbf{u}\) is the \(C_3\) axis unit vector, \(d_{\text{ex}}\) is the constant of antisymmetric exchange for a pair of nearest magnetic ions, \(Z_i\) is the number of nearest magnetic neighbours for the \(i\)th ion and the summation is over all magnetic ions, \(N\), in the volume \(V\).

Taking into account Equation (1) we get:

\[ E_{\text{DM}}^{\text{mix}} = 2 \frac{d_{\text{ex}}}{V n_{\text{mix}}} \sum_{i=1}^{N} Z_i \cdot (M_1, M_2, \mathbf{u}) \]  

(4)

where \(n_{\text{mix}} = N/V\) is the concentration of iron ions in diluted crystals. Expressing in Equation (4) the sublattice magnetizations through the reduced magnetic vectors, see Equation (2), we get:

\[ E_{\text{DM}}^{\text{mix}} = 2 \frac{d_{\text{ex}}}{V n_{\text{mix}}} \sum_{i=1}^{N} Z_i \cdot (m + l, m - l, \mathbf{u}) \]  

(5)

where

\[ M_{\text{mix}} = \frac{1}{2} n_{\text{mix}} \mu_l \]  

(6)

is the sublattice magnetization for diluted crystals. Transforming the triple scalar product, Equation (5) can be rewritten as:

\[ E_{\text{DM}}^{\text{mix}} = D_{\text{DM}}^{\text{mix}} \cdot (l, m, \mathbf{u}) \]  

(7)

where \(D_{\text{DM}}^{\text{mix}}\) is the Dzyaloshinskii-Moriya interaction constant for diluted crystals:

\[ D_{\text{DM}}^{\text{mix}} = \frac{4 d_{\text{ex}} M_{\text{mix}}^2}{V n_{\text{mix}}^2} \sum_{i=1}^{N} Z_j \]  

(8)

Following the approach described by Seleznyova et al. [13], we assume that \(d_{\text{ex}}\) for the pair of nearest magnetic ions does not depend on the degree of diamagnetic dilution. With this assumption, we get:

\[ d_{\text{ex}} = \frac{1}{24} \frac{n D_{\text{DM}}}{M_0^2} \]  

(9)

where \(D_{\text{DM}} = 1.05 \times 10^7 \text{ Jm}^{-3}\) is the Dzyaloshinskii-Moriya constant for iron borate [14]. Thus,

\[ D_{\text{DM}}^{\text{mix}} = \frac{1}{6} \frac{D_{\text{DM}}}{M_0^2} \frac{n M_{\text{mix}}^2}{n_{\text{mix}}} \sum_{i=1}^{N} Z_j \]  

(10)

In order to numerically calculate \(D_{\text{DM}}^{\text{mix}}\), a computer code implementing the summation in Equation (10) has been developed. The random substitution of a part of the magnetic iron ions by the diamagnetic ones has been modelled using the Monte Carlo technique [15]. Figure 2 shows the result of this calculation. As one can see, with decreasing \(x\) \(D_{\text{DM}}^{\text{mix}}\) decreases following a square low.

This behaviour can be explained as follows. Taking into account that for Fe\(_x\)Ga\(_{1-x}\)BO\(_3\), the mean number of nearest magnetic neighbours of a given iron ion is:
1 \sum_{i=1}^{N} Z_i = 6x \quad (11)

and \( n_{\text{mix}} = nx \), using Equations (9) and (6), one can rewrite Equation (10) as:

\[ D_{\text{DM}}^{\text{mix}} = 6n d_{\text{ex}}x^2 = D_{\text{DM}} x^2, \quad (12) \]

thus confirming the numerical calculation, see Figure 2.

![Figure 2](image)

**Figure 2.** Calculated dependence of \( D_{\text{DM}}^{\text{mix}} \times 10^{-6} \) on \( x \) in Fe\(_x\)Ga\(_{1-x}\)BO\(_3\) single crystals (circles). The dashed line is a fitting according to Equation (12).

### 4. Conclusions

The constant of antisymmetric exchange for a pair of nearest magnetic ions, \( d_{\text{ex}} \), is expressed through the Dzyaloshinskii-Moriya constant \( D_{\text{DM}} \) for iron borate FeBO\(_3\). An analytical expression for the Dzyaloshinskii-Moriya constant \( D_{\text{DM}}^{\text{mix}} \) for diluted iron-gallium borates Fe\(_x\)Ga\(_{1-x}\)BO\(_3\) is obtained under the assumption that \( d_{\text{ex}} \) does not change with diamagnetic dilution. In order to model diamagnetically diluted crystal lattice and implement a summation over the nearest neighbours of iron ions, a computer code is put forward. Both numerical and analytical calculations show that the \( D_{\text{DM}}^{\text{mix}} \) vs. \( x \) dependence follows the square-law. The results of the present work will provide the basis for detailed interpretation of experimental results on magnetometry and EMR studies of Fe\(_x\)Ga\(_{1-x}\)BO\(_3\) crystals which are actually in progress.

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