The effects of intra-dimer Dzyaloshinsky–Moriya interaction on the properties of $SrCu_2(BO_3)_2$ in an external magnetic field

The effects of intra-dimer Dzyaloshinsky–Moriya interaction on the properties of $SrCu_2(BO_3)_2$ in an external magnetic field

S Miyahara[1,6], **F Mila**[2], **K Kodama**[3], **M Takigawa**[3], **M Horvatic**[4], **C Berthier**[4], **H Kageyama**[3,5] **and Y Ueda**[3]

[1] Department of Physics, Aoyama Gakuin University, Sagamihara, Kanagawa 229-8558, Japan
[2] Institut de Physique Théorique, Université de Lausanne, CH-1015 Lausanne, Switzerland
[3] Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan
[4] Grenoble High Magnetic Field Laboratory, CNRS and MPI-FKF, 38042 Grenoble, France
[5] Department of Chemistry, Graduate School of Science, Kyoto University,
Kyoto 606-8502, Japan

E-mail: miyahara@phys.aoyama.ac.jp

## Abstract

The two-dimensional spin-gap system $SrCu_2(BO_3)_2$ shows unique physical properties due to the strong quantum fluctuations that follow from its low-dimensionality and its very special topology. The magnetic properties of this material, e.g. dimer singlet ground state, an almost localized triplet and magnetization plateaux, are well described by a spin-$1/2$ antiferromagnetic Heisenberg model on the orthogonal dimer lattice. Recently, an unusual angular dependence of the shifts at the B sites with the external magnetic field rotated in the (110) plane, which indicates the presence of a field-induced staggered moment, has been observed by nuclear magnetic resonance. Such a behaviour cannot be explained by the Heisenberg model alone. We include an intra-dimer Dzyaloshinsky–Moriya interaction as well as an anisotropic gyromagnetic tensor in the orthogonal dimer model, and we show that the observed field-induced staggered moment can be very well described with physically reasonable values of the parameters.

1. Introduction

After the re-discovery of the two-dimensional spin-gap system $SrCu_2(BO_3)_2$ [1], much experimental and theoretical research has been done on this material [2]. The unique magnetic properties of this material, (1) a dimer singlet ground state, (2) an almost localized triplet

---

[6] Author to whom any correspondence should be addressed.
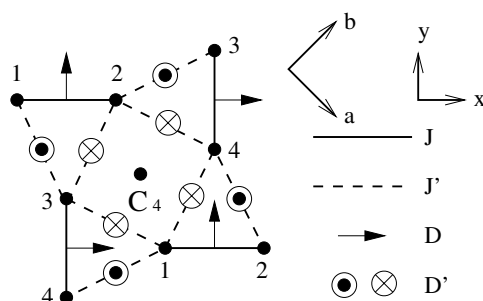
Figure 1. The orthogonal dimer model for $SrCu_2(BO_3)_2$. The unit cell contains four inequivalent spin sites. The direction of the possible Dzyaloshinsky–Moriya interactions are also indicated.

excitation and (3) magnetization plateaux, are well described by a spin-1/2 antiferromagnetic Heisenberg model on the orthogonal dimer lattice (figure 1). However, some features cannot be explained by the Heisenberg model alone. For example, a splitting of the lowest excited triplet state, which has been observed by electron spin resonance (ESR) [3] and inelastic neutron scattering [4] experiments, can be well described by including Dzyaloshinsky–Moriya interactions on next nearest neighbour bonds D' [4]. Recently, an unusual angular dependence of shifts at the B sites, with an external magnetic field rotated in the (110) plane, has been observed by nuclear magnetic resonance (NMR) [5]. This is another property which cannot be explained by the Heisenberg model alone. In this paper, we include a Dzyaloshinsky–Moriya interaction on nearest neighbour bonds D and an anisotropic gyromagnetic tensor in the orthogonal dimer Heisenberg model, and discuss their effects in the presence of an external magnetic field.

2. NMR results

We have measured anisotropic shifts at the B sites with the external magnetic field H in the plane including the c-axis and the x direction. The angular dependence of the shift K_1 at several temperatures and for a magnetic field of 7 T are shown in figure 2(a). The shifts K_i ($i = 1, 2, 3$ and 4, see figure 1) reflect the nature of the local magnetization on Cu sites because of the nearest neighbour hyperfine couplings between B and Cu sites (we may neglect the other hyperfine couplings as the first approximation). If the ground state is a product of singlets, any spin response should be very small at $T = 2.5$ K, which is much lower than the estimated value of the spin gap 35 K. However, the data indicate that the field induces staggered magnetization. The evidence for staggered magnetization can be gained most clearly from the temperature dependence of the angular variation of $(K_1 - K_2)/2$ (figure 2(b)). The amplitude of the angular variation $(K_1 + K_2)/2$ and $(K_3 + K_4)/2$ approximately follow the temperature dependence of the bulk susceptibility. However the amplitude $(K_1 - K_2)/2$ does not decrease as rapidly as other components and has a finite amplitude around $T = 0$ K. Note that the amplitude of the angular variation of $(K_1 + K_2)/2$ and $(K_3 + K_4)/2$ is strongly coupled to the uniform magnetization through the diagonal component of the hyperfine tensor. On the other hand, $(K_1 - K_2)/2$ is coupled to the staggered components.

3. Model Hamiltonian

In Cu-benzoate [6], a field-induced staggered magnetization has previously been proposed by a theory based on an effective staggered field resulting from a staggered gyromagnetic tensor
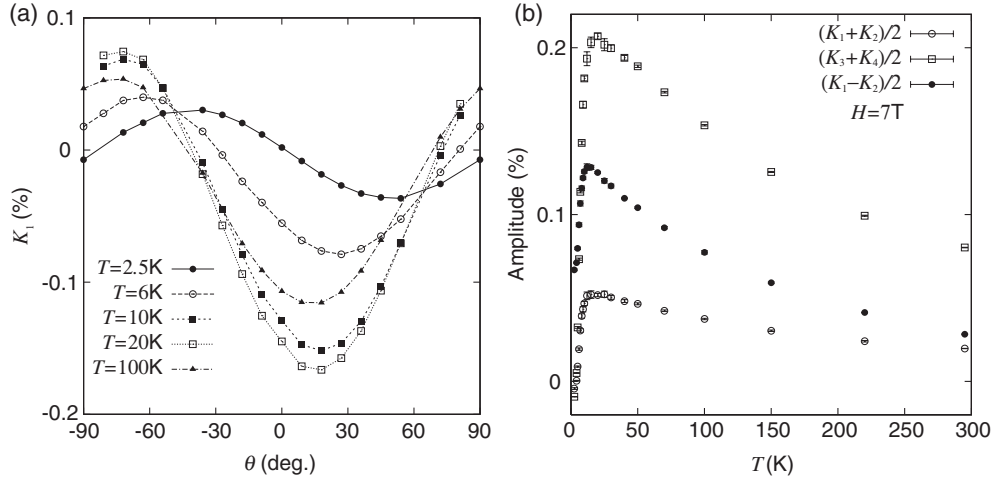
Figure 2. (a) The anisotropic shifts at the ^{11}B sites with the magnetic field of 7 T rotated in the (110) plane at $T = 2.5, 6, 10, 20$ and 100 K. The shifts K_1 reflect a spin state at site 1. Curves are guides to the eyes. (b) The temperature dependence of the angular variation of $(K_1 + K_2)/2$, $(K_3 + K_4)/2$ and $(K_1 - K_2)/2$.

and a Dzyaloshinsky–Moriya interaction [7, 8]. In the same spirit as in [7, 8], we include a staggered gyromagnetic tensor and a Dzyaloshinsky–Moriya interaction in the orthogonal dimer model. Note, however, that there are important differences between the two systems. In particular, Cu-benzoate is an antiferromagnetic spin chain with a magnetic ground state, and in which a magnetic field opens a gap, whereas SrCu₂(BO₃)₂ is a two-dimensional system which already has a spin gap in zero field.

The model Hamiltonian of the orthogonal dimer model with Dzyaloshinsky–Moriya interactions and staggered gyromagnetic tensors is written as

$$H = J \sum_{n.n.} \mathbf{s}_i \cdot \mathbf{s}_j + J' \sum_{n.n.n.} \mathbf{s}_i \cdot \mathbf{s}_j + D_y \sum_{\text{A-dimer}} (s_1^z s_2^x - s_1^x s_2^z) + D_x \sum_{\text{B-dimer}} (s_3^y s_4^z - s_3^z s_4^y)$$
$$- \sum_{\text{unitcell}} ((\hat{g}_1 \mathbf{H}) \cdot \mathbf{s}_1 + (\hat{g}_2 \mathbf{H}) \cdot \mathbf{s}_2 + (\hat{g}_3 \mathbf{H}) \cdot \mathbf{s}_3 + (\hat{g}_4 \mathbf{H}) \cdot \mathbf{s}_4). \quad (1)$$

In the model, there are two inequivalent dimers, horizontal (A-dimer) and perpendicular (B-dimer) ones. Thus the unit cell contains four inequivalent spin sites, i.e. four inequivalent gyromagnetic tensors. If the CuBO₃ planes were flat and had mirror symmetry, the J-bonds would have an inversion centre and the Dzyaloshinsky–Moriya interaction would not be allowed. However, in the real material at low temperatures (<395 K), there is a buckling in the plane, and the mirror symmetry with respect to the *ab* plane is lost [9, 10]. Thus the centre of the J bonds is not an inversion symmetry point and the components of the Dzyaloshinsky–Moriya interaction shown in figure 1 can exist [11].

The Dzyaloshinsky–Moriya interactions D_x and D_y are connected to each other by symmetry operations, namely the rotation C_4 (see the figure) followed by the mirror I (*ab*-plane), leading to the relation $D_x = -D_y$. The gyromagnetic tensor on site 1 is written as

$$\hat{g}_1 = \begin{pmatrix} g_\xi \cos^2 \varphi + g_\zeta \sin^2 \varphi & 0 & (g_\xi - g_\zeta) \cos \varphi \sin \varphi \\ 0 & g_\eta & 0 \\ (g_\xi - g_\zeta) \cos \varphi \sin \varphi & 0 & g_\xi \sin^2 \varphi + g_\zeta \cos^2 \varphi \end{pmatrix} = \begin{pmatrix} g_x & 0 & g_s \\ 0 & g_y & 0 \\ g_s & 0 & g_z \end{pmatrix} \quad (2)$$

based on a symmetry analysis of the crystallographic structure [9, 10]. We set the x, y, z coordinates such that z is along the c-axis and x is along the [110] direction (see figure 1).

The diagonal elements of the g-tensor with respect to its principal axes are denoted by g_ξ, g_η and g_ζ. One of the principal axes is normal to the mirror plane (the (110) plane) and one other principal axis is tilted slightly from the z-axis. Here the angle φ is the angle between the ζ-axis and the z-axis. The other three tensors are related to the tensor of equation (2) by symmetry operations, and the results are

$$\hat{g}_2 = \begin{pmatrix} g_x & 0 & -g_s \\ 0 & g_y & 0 \\ -g_s & 0 & g_z \end{pmatrix}, \qquad \hat{g}_3 = \begin{pmatrix} g_y & 0 & 0 \\ 0 & g_x & -g_s \\ 0 & -g_s & g_z \end{pmatrix}, \qquad \hat{g}_4 = \begin{pmatrix} g_y & 0 & 0 \\ 0 & g_x & g_s \\ 0 & g_s & g_z \end{pmatrix}.$$
(3)

From ESR experiments, the diagonal terms can be estimated to be $g_x = g_y = 2.05$ and $g_z = 2.28$ [12]. The off-diagonal elements g_s are staggered, i.e. they are opposite on sites 1 and 2, respectively 3 and 4. Since it is difficult to estimate the value for g_s directly from experiments, we treat it as a parameter in the model.

4. Ground state

Without Dzyaloshinsky–Moriya interactions and staggered gyromagnetic tensors, the ground state of the orthogonal dimer Heisenberg model is the product of the dimer singlets [13, 14],

$$|s\rangle = \prod_i |s\rangle_i = \prod_i \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle_i - |\downarrow\uparrow\rangle_i).$$
(4)

Here i denotes nearest-neighbour bonds.

To clarify the effects of Dzyaloshinsky–Moriya interactions and staggered gyromagnetic tensors, we performed a perturbation calculation in the limit $D_x/J, g_s H/J \ll g_x H/J \ll 1$. Since the main magnetic properties can be well described by the Heisenberg model, it is natural to assume that D_x and $g_s H$ are much smaller than J. In the presence of an external magnetic along the [110] direction $\mathbf{H} = (H \sin\theta, 0, H \cos\theta)$, the expectation values $\langle S_i^x \rangle$, $\langle S_i^y \rangle$ and $\langle S_i^z \rangle$ are given to first order in D_y/J and $g_s H/J$ by:

$$\langle S_1^x \rangle = -\frac{g_s H \cos\theta}{\Delta} + \frac{D_y g_z H \cos\theta}{2\Delta^2}$$
(5)

$$\langle S_2^x \rangle = -\langle S_1^x \rangle$$
(6)

$$\langle S_1^z \rangle = \frac{g_s H \sin\theta}{\Delta} - \frac{D_y g_x H \sin\theta}{2\Delta^2}$$
(7)

$$\langle S_2^z \rangle = -\langle S_1^z \rangle$$
(8)

$$\langle S_3^y \rangle = -\frac{g_s H \cos\theta}{\Delta} + \frac{D_y g_z H \cos\theta}{2\Delta^2}$$
(9)

$$\langle S_4^y \rangle = -\langle S_3^y \rangle.$$
(10)

It is clear that the staggered magnetization is induced by the external fields. The expectation values of other components, $\langle S_1^y \rangle$, $\langle S_2^y \rangle$, $\langle S_3^x \rangle$, $\langle S_3^z \rangle$, $\langle S_4^x \rangle$ and $\langle S_4^z \rangle$, vanish.

Finally, let us consider the case $\mathbf{H} \sim (0, 0, H_c)$. Here H_c is the critical field at which the spin gap would collapse in the absence of the Dzyaloshinsky–Moriya interactions and staggered gyromagnetic tensor. In this field region, we may take the lowest energy states: $|s\rangle$ (equation (4)) and $|e_1(i)\rangle$, where

$$|e_1(i)\rangle = |t_1\rangle_i \prod_{j \neq i} |s\rangle_j.$$
(11)

Although one triplet states (equation (11)) are degenerate, only the matrix element between the ground state and a Fourier transformation of equation (11) with $\mathbf{q} = 0$ can be non-zero, since

the ground state for the Heisenberg model $|s\rangle$ is uniform. Thus in the perturbation calculations in the third order for J'/J and first order for D_y/J and $g_s H/J$, the ground state on the A-dimer bond is given by solving the 2×2 matrix:

$$\begin{pmatrix} 0 & \frac{D_y}{2} + \frac{g_s}{\sqrt{2}} H \\ \frac{D_y}{2} + \frac{g_s}{\sqrt{2}} H & J\left(1 - \left(\frac{J'}{J}\right)^2 - \frac{1}{2}\left(\frac{J'}{J}\right)^3\right) - g_z H \end{pmatrix}. \tag{12}$$

The state on the A-dimer bond is nothing but the ground state for an isolated dimer with a renormalized spin gap. Note that a state on the B-dimer is also obtained through a similar calculation. Thus the gap persists even at H_c and the magnetization can be finite below H_c. This picture can be a good approximation for the fields around H_c, where we can neglect the mixing of the bound state of two triplets to the ground state. These features are qualitatively consistent with the results observed in the experiments [3, 12, 15, 16].

5. Conclusion

Including an intra-dimer Dzyaloshinsky–Moriya interaction D and staggered gyromagnetic tensors, one can qualitatively explain the staggered magnetization which was observed in NMR experiments. Let us comment on the effects of Dzyaloshinsky–Moriya interactions on next-nearest-neighbour bond D', which was included to explain the magnetic behaviours for lowest triplet excitations [4]. If we apply the D' term to the ground state, two of the singlet states are promoted to triplets. Thus a non-zero expectation value for $\langle S_i^\alpha \rangle$ ($\alpha = x, y, z$) only appears at second order in D'/J, and it would be much smaller than that deduced from NMR experiments since $D'/J \sim 0.02$. In contrast, $\langle S_i^\alpha \rangle$ is proportional to D, and if we assume that $D \sim D'$, the magnitude of the calculated expectation values is consistent with the observed one. On the other hand, in the lowest branches of excitations the effects of D' are more important than those of D. If we apply the D' term to the state (11), the triplet can hop to the neighbouring bonds, and the magnitude of the splitting observed in the lowest branch of the excitation is proportional to D', whereas the D term would lead to a splitting proportional to D^2. These results indicate that, although D and D' have in principle similar effects since they both break $SU(2)$ symmetry, D dominates the behaviour of the ground state while D' is crucial to understanding the lowest branch of the triplet excitations.

Acknowledgments

References

[1] Kageyama H, Yoshimura K, Stern R, Mushnikov N V, Onizuka K, Kato M, Kosuge K, Slichter C P, Goto T and Ueda Y 1999 Exact dimer ground state and quantized magnetization plateaus in the two-dimensional spin system SrCu$_2$(BO$_3$)$_2$ *Phys. Rev. Lett.* **82** 3168–71
[2] Miyahara S and Ueda K 2003 Theory of the orthogonal dimer Heisenberg spin model for SrCu$_2$(BO$_3$)$_2$ *J. Phys.: Condens. Matter* **15** R327–66
[3] Nojiri H, Kageyama H, Onizuka K, Ueda Y and Motokawa M 1999 Direct observation of the multiple spin gap excitations in two-dimensional dimer system SrCu$_2$(BO$_3$)$_2$ *J. Phys. Soc. Japan* **68** 2906–9
[4] Cépas O, Kakurai K, Regnault L P, Boucher J P, Ziman T, Aso N, Nishi M, Kageyama H and Ueda Y 2001 Dzyaloshinski–Moriya interaction in the 2d spin gap system SrCu$_2$(BO$_3$)$_2$ *Phys. Rev. Lett.* **87** 167205
[5] Kodama K, Arai K, Takigawa M, Kageyama H and Ueda Y 2003 Spin correlation and field induced staggered magnetization in the 2d orthogonal dimer spin system SrCu$_2$(BO$_3$)$_2$ *J. Magn. Magn. Mater.* at press

[6] Dender D C, Hammar P R, Reich D H, Broholm C and Aeppli G 1997 Direct observation of field-induced incommensurate fluctuations in a one-dimensional $S = 1/2$ antiferromagnet *Phys. Rev. Lett.* **79** 1750–3

[7] Oshikawa M and Affleck I 1997 Field-induced gap in $s = 1/2$ antiferromagnetic chains *Phys. Rev. Lett.* **79** 2883–6

[8] Affleck I and Oshikawa M 1999 Field-induced gap in Cu benzoate and other $s = \frac{1}{2}$ antiferromagnetic chains *Phys. Rev.* B **60** 1038–56

[9] Smith R W and Keszler D A 1991 Synthesis, structure, and properties of the orthoborate $SrCu_2(BO_3)_2$ *J. Solid State Chem.* **93** 430–5

[10] Sparta K, Redhammer G J, Roussel P, Heger G, Roth G, Lemmens P, Ionescu A, Grove M, Güntherodt G, Hüning F, Lueken H, Kageyama H, Onizuka K and Ueda Y 2001 Structural phase transition in the 2d spin dimer compound $SrCu_2(BO_3)_2$ *Eur. Phys. J.* B **19** 507–16

[11] Moriya T 1960 Theory of magnetism of NiF_2 *Phys. Rev.* **117** 635–47

[12] Nojiri H, Asano T, Ajiro Y, Kageyama H, Ueda Y and Motokawa M 2001 High-field ESR on quantum spin systems *Physica* B **294–295** 14–9

[13] Shastry B S and Sutherland B 1981 Exact ground state of a quantum mechanical antiferromagnet *Physica* B **108** 1069–70

[14] Miyahara S and Ueda K 1999 Exact dimer ground state of the two dimensional Heisenberg spin system for $SrCu_2(BO_3)_2$ *Phys. Rev. Lett.* **82** 3701–4

[15] Onizuka K, Kageyama H, Narumi Y, Kindo K, Ueda Y and Goto T 2000 1/3 magnetization plateau in $SrCu_2(BO_3)_2$—stripe order of excited triplets *J. Phys. Soc. Japan* **69** 1016–8

[16] Tsujii H, Rotundu R C, Andraka B, Takano Y, Kageyama H and Ueda Y 2003 *Preprint* cond-mat/0301509