Origin of magnetic anisotropy in the spin ladder compound \((\text{C}_5\text{H}_{12}\text{N})_2\text{CuBr}_4\)

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The \(S = 1/2\) spin ladder compound \((\text{C}_5\text{H}_{12}\text{N})_2\text{CuBr}_4\) (BPCB) is studied by means of high-resolution inelastic neutron scattering. In agreement with previous studies we find a band of triplet excitations with a spin gap of \(\sim 0.8\) meV and a bandwidth of \(\sim 0.6\) meV. In addition, we observe a distinct splitting of the triplet band of \(50(1)\) \(\mu\)eV or \(40(2)\) \(\mu\)eV at the band minimum or maximum, respectively. By comparison to a strong coupling expansion calculation of the triplet dispersion for a spin ladder with anisotropic exchange, weakly anisotropic leg interactions are identified as the dominant source of magnetic anisotropy in BPCB. Based on these results, we discuss the nature of magnetic exchange anisotropy in BPCB and in related transition-metal insulators.

I. INTRODUCTION

The Heisenberg model is arguably the most important construct in quantum magnetism. Experimentally, it is usually studied in transition metal oxides and metal-organic salts. Unfortunately, symmetry-breaking magnetic anisotropy is unavoidable in such systems. Even when weak compared to exchange interactions, it can have a significant influence on the magnetic and dynamical properties. In understanding these, existing elaborate microscopic theories are usually dismissed in favor of minimalistic empirical descriptions using ad hoc parameters. How justified is such an approach? Here we present a case study: the effect of very weak magnetic anisotropy on spin excitations in the almost ideal Heisenberg quantum spin ladder compound \((\text{C}_5\text{H}_{12}\text{N})_2\text{CuBr}_4\) (BPCB). Exceptionally high resolution inelastic neutron scattering data allow an in-detail investigation of the anisotropy’s microscopic origins.

The paper proceeds as follows: In Sec. II we describe the compound BPCB and we present our inelastic neutron scattering data. In Sec. III we calculate the triplet dispersion of a spin ladder with exchange anisotropy in a strong coupling expansion. In Sec. IV we interpret our data and discuss the origin of magnetic anisotropy in BPCB. These findings we compare to the theory of anisotropic superexchange and we discuss how the present case study may guide our understanding of magnetic anisotropy in related compounds.

II. EXPERIMENT AND RESULTS

A. The compound \((\text{C}_5\text{H}_{12}\text{N})_2\text{CuBr}_4\): crystal structure and magnetic interaction pathways

The compound \((\text{C}_5\text{H}_{12}\text{N})_2\text{CuBr}_4\), bis-piperidinium copper bromide or BPCB for short crystalizes in a monoclinic crystal structure with space group \(\text{P}2_1/c\) and room temperature lattice parameters \(a = 8.49\), \(b = 17.22\), \(c = 12.38\) Å, and \(\beta = 99.3^\circ\). The spin ladders are formed by the magnetic \(\text{Cu}^{2+}\) cations carrying a spin \(S = 1/2\) linked by super-exchange bridges via

![Schematic of the crystal structure of BPCB. The spin ladders are formed by the magnetic \(\text{Cu}^{2+}\) cations and linking \(\text{Br}^-\) anions. Crystallographic centers of inversion symmetry are marked by green dots. Anisotropic Dzyaloshinskii-Moriya interactions, by symmetry are only allowed on the ladder legs and possible DM vectors \(\text{D}\) are sketched as blue arrows on these bonds. Note that for this monoclinic structure, in both plots the third axis (projection axis) is not perpendicular to the plane of the figure.](image)

\(\text{Br}^-\) anions. The ladders run along the crystallographic \(a\) axis and are well separated by non-magnetic organic piperidinium ions as depicted in Fig. II.1

There are two crystallographically equivalent spin ladders in BPCB, related by the glide plane symmetry of the \(\text{P}2_1/c\) space group. Furthermore, the center of each ladder rung corresponds to a crystallographic center of inversion symmetry. For this reason, on the ladder rungs, only symmetric exchange is possible whilst for the ladder legs there are no symmetry restrictions and both symmetric and antisymmetric exchange are in principle allowed. On the ladder leg, the antisymmetric exchange contribution is parameterized by a Dzyaloshinskii-Moriya (DM) vector \(\text{D}\). These DM vectors are uniform within every ladder leg and anti-aligned between the two legs of each ladder. The DM vectors of the two crystallographically equivalent ladders are again
related by the reflection of the glide plane symmetry as sketched in Fig. 1. Besides these relations, the DM vectors may point in any direction and are not further constrained by symmetry. Similarly, the $g$-tensors for the magnetic moments residing on the different Cu$^{2+}$ ions show different principal axes for the two types of ladders. Thus, in an applied magnetic field, the two types of ladders become inequivalent, except for special orientations of the magnetic field. In the present study, however, we exclusively focus on zero-field properties where the two ladders remain equivalent.

### B. Inelastic neutron scattering

Neutron scattering experiments were performed on four fully deuterated single crystals of total mass 2.07 g, co-aligned to better than 1° effective mosaic spread. The measurements were carried out at the LET cold neutron time-of-flight spectrometer at the ISIS facility, UK. The sample was mounted on a $^3$He-$^4$He dilution refrigerator with the crystallographic $b$ axis vertical. Making use of repetition rate multiplication, data was collected simultaneously using neutrons of incident energies $E_i = 1.35, 2.20, 4.20$ and 11.0 meV. For these configurations we find an approximately Gaussian energy resolution of 19, 36, 97 and 410 $\mu$eV at FWHM, respectively, for elastic scattering ($h\omega = 0$) and improving towards higher energy transfer.

### C. Experimental results

An overview of the neutron scattering data collected for $E_i = 2.2$ and 1.35 meV at a temperature of 0.35 K is presented as false color intensity plots in Fig. 2. For this quasi-one-dimensional spin ladder system, the measured scattering intensity is plotted versus the reduced wave-vector transfer parallel to the ladder $q|| = Q \cdot a$ and integrated fully along the non-dispersive $b^*$ and $c^*$ directions.

We observe a resolution-limited band of excitations with an excitation gap of $\sim 0.8$ meV, a bandwidth of $\sim 0.6$ meV and a distinct splitting at the band extrema. Cuts through both data sets at the band extrema are shown in Fig. 3. These correspond to the data of Fig. 2 integrated in thin slices of $q||/2\pi = -0.5 \pm 0.02$ and $q||/2\pi = 0 \pm 0.03$, respectively (the dispersion is less curved at the maximum allowing to integrate a wider slice for improved counting statistics). To these cuts we fit two Gaussian peaks of equal width and a linear background shown as a solid lines in Fig. 3. We find a splitting of $\Delta_{\text{min}} = 50(1) \mu$eV at the band minimum and $\Delta_{\text{max}} = 40(2) \mu$eV at the band maximum.
III. TRIPLET DISPERSION FOR A SPIN LADDER WITH EXCHANGE ANISOTROPY

A. Isotropic Heisenberg ladder

For an isotropic spin $S = 1/2$ ladder we denote the rung and leg Heisenberg exchange constants as $J_\perp$ and $J_\parallel$, respectively. In the limit of $\lambda = J_\parallel / J_\perp \ll 1$, the spin ladder can be understood as a 1D array of weakly interacting spin dimers. For $\lambda = 0$ the groundstate corresponds to a product state of singlets and the low energy excitations are triplet excitations. In the presence of weak leg coupling, these excitations become mobile and for small $\lambda$ their dispersion can be calculated in a strong coupling expansion\cite{ref18}. For an isotropic ladder with Hamiltonian

$$\mathcal{H} = J_\perp \left( \mathcal{H}^\perp + \lambda \mathcal{H}^\parallel \right), \quad \mathcal{H}^\perp = \sum_{R=1}^{L} \mathbf{S}_{R,1} \cdot \mathbf{S}_{R,2}, \quad \mathcal{H}^\parallel = \sum_{R=1}^{L} \sum_{\alpha=1}^{2} \mathbf{S}_{R,\alpha} \cdot \mathbf{S}_{R+1,\alpha},$$

up to $3^{rd}$ order in $\lambda$, the following dispersion is obtained in Ref.\cite{ref18} degenerate for the three $\sigma = \{+, 0, -\}$ triplet branches:

$$\epsilon_\sigma(k)/J_\perp = 1 + \gamma \cos(k) + \frac{\gamma^2}{4} \left[ 3 - \cos(2k) \right] + \frac{\gamma^3}{8} \left[ 3 - 2 \cos(k) - 2 \cos(2k) + \cos(3k) \right].$$

B. Anisotropic rung interactions

In addition to the isotropic rung and leg couplings, here Ising-type anisotropy on the ladder rungs is considered. We choose the $z$ axis parallel the Ising axis and consider the Hamiltonian

$$\mathcal{H} = J_\perp \left( \mathcal{H}^\perp + \lambda \mathcal{H}' \right), \quad \mathcal{H}' = \mathcal{H}^\parallel + C \mathcal{H}_{\text{Rung,Ising}}, \quad \mathcal{H}_{\text{Rung,Ising}} = \sum_{R=1}^{L} S_{R,1}^z S_{R,2}^z,$$

where the Ising anisotropy on the ladder rungs is parameterized by $C$. Using the same strong coupling expansion as applied to the isotropic case in Ref.\cite{ref18} up to $3^{rd}$ order in $\lambda$ we find

$$\epsilon_0(k)/J_\perp = 1 + \lambda \cos(k) + \frac{\lambda^2}{4} \left[ 3 - \cos(2k) \right] + \frac{\lambda^3}{8} \left[ 3 - 2C - 2 \cos(k) - 2 \cos(2k) + \cos(3k) \right]$$

$$\epsilon_\pm(k)/J_\perp = 1 + \lambda \cos(k) + \frac{\lambda^2}{4} \left[ 3 - \cos(2k) \right] + \frac{\lambda^3}{8} \left[ 3 - 2C - 2 \cos(k) + (C - 2) \cos(2k) + \cos(3k) \right].$$

C. Anisotropic leg interactions

Finally, we consider anisotropic exchange on the ladder legs with both symmetric and anti-symmetric contributions. Assuming a center of inversion symmetry on the ladder runs, we consider a uniform DM vector $+\mathbf{D}$ on one leg and $-\mathbf{D}$ on the other leg. Furthermore we include Ising anisotropy of independent magnitude but pointing in the same direction as $\mathbf{D}$. We choose the $z$ axis parallel to $\mathbf{D}$ and consider

$$\mathcal{H} = J_\perp \left( \mathcal{H}^\perp + \lambda \mathcal{H}' \right), \quad \mathcal{H}' = \mathcal{H}^\parallel + B \mathcal{H}_{\text{Leg,DM}} + A \mathcal{H}_{\text{Leg,Ising}}$$

$$\mathcal{H}_{\text{Leg,DM}} = \sum_{R=1}^{L} \sum_{\alpha=1}^{2} (-1)^\alpha \mathbf{e}_z \cdot (\mathbf{S}_{R,\alpha} \times \mathbf{S}_{R+1,\alpha}), \quad \mathcal{H}_{\text{Leg,Ising}} = \sum_{R=1}^{L} \sum_{\alpha=1}^{2} S_{R,\alpha}^z S_{R+1,\alpha}^z$$

Up to $3^{rd}$ order in $\lambda$ we find the following triplet dispersion:

$$\epsilon_0(k)/J_\perp = 1 + \lambda(1 + A) \cos(k) + \frac{\lambda^2}{4} \left[ 3 - 4B^2 + 2A + A^2 + 4B^2 \cos(k) - (1 + 2A + A^2) \cos(2k) \right]$$

$$+ \frac{\lambda^3}{8} \left[ 3 + 3A - \left( 2 + 4B^2 + 4A + 4AB^2 + 3A^2 + A^3 \right) \cos(k) \right.$$

$$- \left( 2 + 2A - 4B^2 - 4AB^2 \right) \cos(2k) + \left( 1 + 3A + 3A^2 + A^3 \right) \cos(3k) \right]$$

$$\epsilon_\pm(k)/J_\perp = 1 + \lambda \cos(k) + \frac{\lambda^2}{4} \left[ 3 - 2B^2 + 2A + A^2 - \cos(2k) \right]$$

$$+ \frac{\lambda^3}{8} \left[ 3 + 3A - \left( 2 + 12B^2 + A + \frac{A^2}{2} \right) \cos(k) - \left( 2 + 2B^2 + 2A + 2AB^2 \right) \cos(2k) + \frac{1}{8} \cos(3k) \right].$$
IV. DISCUSSION

A. Discussion of experimental results

The experimental data shown in Fig. 2 closely resembles previously published results on BPCB with dispersive triplet excitations. In these experiments the resolution was not sufficient to detect the small band splitting and those data were very well described using a model of an isotropic spin ladder with splitting and those data were very well described using a model of an isotropic spin ladder with

\[ J_{\perp}/k_B = 12.7(1) \text{ K and } J_{\parallel}/k_B = 3.54(3) \text{ K}. \]

Only with the superior resolution of the present experiment, the small splitting of the triplet band is observed.

In our data, perpendicular to the one-dimensional ladders, no dispersion is observed within the experimental resolution. This validates BPCB as an exceptionally one-dimensional ladder compound and it corroborates, that the observed band splitting is inherent to the spin ladders and is not related to residual 3D interactions. In addition, inter-ladder interactions were previously estimated on the order of a few 10s of mK (a few µeV) – much smaller than the observed band splitting.

A previous ESR study on BPCB found clear signatures of magnetic anisotropy, of exactly the same magnitude as the band splitting observed in the present experiments. By a careful study of the angular dependence of the ESR signal, an anisotropy axis tilted approx. 50° from the b axis in the (b, c*) plane could be identified as a special direction. However, the origin of this anisotropy remained unclear.

One source of magnetic anisotropy are dipolar interactions present in all materials. For classical magnetic moments at the shortest Cu-Cu distance found in BPCB, dipolar interactions are estimated as \( J_{\text{dipolar}}/k_B \approx 6 \text{ mK (}< 1 \text{ µeV)} \), much smaller than the observed band splitting. Dipolar interactions clearly can be ruled out as the primary cause of magnetic anisotropy in BPCB. Since for the \( S = 1/2 \text{ Cu}^{2+} \) ions there cannot be any single-ion anisotropy, exchange anisotropy must be the dominant cause of the observed small splitting of the triplet band. Before further analyzing our data in this regard, we first discuss the nature of anisotropic super-exchange interactions in the following section.

B. Anisotropic super-exchange

Most generally, the effective exchange interaction between two spins takes the form \( H_{1,2} = S_1 \Gamma S_2 \) where \( \Gamma \) is a tensor. Decomposing \( \Gamma \) into its symmetric and anti-symmetric parts and choosing a basis such that the symmetric part is diagonal, this can be written as

\[ H_{1,2} = J \mathbf{S}_1 \cdot \mathbf{S}_2 + D \cdot (\mathbf{S}_1 \times \mathbf{S}_2) + \sum_{\alpha=x,y,z} G^\alpha S_1^\alpha S_2^\alpha. \]  

Here, \( J \) is the isotropic (Heisenberg) exchange. The so-called Dzyaloshinskii-Moriya (DM) vector \( D \) quantifies the anti-symmetric exchange and \( G^\alpha (\alpha = x, y, z) \) determine the symmetric (Ising) exchange anisotropy where \( \sum_{\alpha} G^\alpha = 0 \).

Including spin-orbit coupling (SOC) into Anderson’s theory of super-exchange, indeed all these contributions are obtained. However, in this setting, symmetric and anti-symmetric interactions are not independent and for a single bond, to a good approximation the super-exchange interaction Hamiltonian reads

\[ H_{1,2} = \left( J - \frac{|D|^2}{4J} \right) S_1 S_2 + D \cdot (S_1 \times S_2) + \frac{1}{2J} (\mathbf{D} \cdot \mathbf{S}_1)(\mathbf{D} \cdot \mathbf{S}_2). \]

Here a single vector \( \mathbf{D} \) defines both the symmetric and antisymmetric exchange. This expression, although not obvious, is fully invariant under spin rotations. While the term \( \mathbf{D} \cdot (\mathbf{S}_1 \times \mathbf{S}_2) \) acts to confine the spins to a plane perpendicular to the vector \( \mathbf{D} \), the Ising contribution acts to align the spins with \( \mathbf{D} \). Thus, for a single bond, anisotropic super-exchange will never single out a particular direction. Nonetheless, for multiple connected bonds, frustration of the \( \mathbf{D} \) vectors may still beak spin rotation symmetry and lead to the appearance of anisotropy.

Besides this mechanism, considering multiple orbitals of the ligands transmitting the super-exchange interactions, in the presence of SOC may result in additional sources of exchange anisotropy, beyond Eq. [11]. However, these additional effects have been argued to be much smaller in magnitude.

This leaves us with the following situation: When considering super-exchange interactions, to first approximation symmetric and antisymmetric anisotropy contributions always come together pointing in the same direction and at a fixed ratio. In this case, if the center of a bond corresponds to a center of inversion symmetry, not only is antisymmetric exchange anisotropy forbidden by symmetry as is commonly known, but we also expect symmetric exchange anisotropy to vanish. Only additional mechanisms may then lead to Ising-type anisotropy which is in principal allowed on a bond with inversion symmetry.

C. Exchange anisotropy in BPCB

For the compound BPCB with centers of inversion symmetry on the ladder rungs, we consider the following cases compatible with the crystallographic symmetry:

a) Ising anisotropy on the ladder rungs
b) Ising anisotropy on the ladder legs
c) DM anisotropy on the ladder legs
d) Both DM and Ising anisotropy on the ladder legs as predicted for simple super-exchange (Eq. [11]).

Using the results of Sec. [III] for these four cases, the triplet dispersion is plotted in Fig. 3. For these plots, we have used the Heisenberg exchange constants \( J_{\perp}/k_B = 12.7 \text{ K and } J_{\parallel}/k_B = 3.54 \text{ K} \) approximately describing
FIG. 4. Triplet dispersion for a spin ladder with exchange anisotropy: a) Ising anisotropy on the ladder rungs, b) Ising anisotropy on the legs, c) DM anisotropy on the legs, and d) both Ising and DM anisotropy on the ladder legs, in the ratio as predicted for a simple super-exchange mechanism (Eq. 11). For these plots, isotropic $J_\perp/k_B = 12.7$ K and $J_\parallel/k_B = 3.54$ K were used, approximately describing the ladders in BPCB. Further, in all four cases the anisotropy parameter was chosen such that the triplet splitting at the band minimum amounts to 50 $\mu$eV. In all plots, the blue line shows the doubly degenerate $\epsilon_{\pm}(k)$ dispersion and the orange line shows the $\epsilon_0(k)$ triplet.

BPCB\cite{13} In all cases the anisotropy parameter was chosen such that the triplet splitting at the band minimum amounts to 50 $\mu$eV.

Comparing the dispersion calculated for Ising-type exchange anisotropy on the ladder rungs (Fig. 4a) to the data of Fig. 2, we can clearly rule out this case as the dominant source of anisotropy in BPCB. The data qualitatively disagree with the calculated dispersion. For the case of anisotropic leg exchange, all three types of anisotropy considered give qualitatively the same dispersion (Fig. 4b,c,d): There is a doubly degenerate band with a smaller bandwidth and a non-degenerate band with larger bandwidth. In all cases the calculated anisotropy splitting is slightly larger at the band minimum than at the band maximum, just as in our data\cite{22}.

In Fig. 5 we show the triplet dispersion extracted from our data. These points were obtained from Gaussian fits to constant-$q_\parallel$-cuts, similar to the ones shown in Fig. 2. The vertical bars denote the width of the observed peaks. Fitting the calculated dispersions with anisotropic leg exchange (cases b,c,d) to this data, we find excellent agreement for all three cases. Indeed, the three calculated dispersions are so similar, that from our data, it is impossible to determine which type of exchange anisotropy on the ladder legs actually causes the observed band splitting in BPCB. As an example, in Fig. 5 the solid line shows the dispersion calculated for anisotropic exchange on the ladder legs given by Eq. 11 (case d) with fitted parameters

$$J_\perp/k_B = 12.77(1) \text{ K,}$$
$$J_\parallel/k_B = 3.55(1) \text{ K,}$$
$$D_\parallel/k_B = 1.44(2) \text{ K.}$$

In the parametrization of the Hamiltonian of Sec. III C, the coefficients are $\tilde{J}_\parallel = J_\parallel - D_\parallel/(4J_\parallel)$, $\lambda = J_\parallel/J_\perp$, $A = D_\parallel/(2J_\parallel \tilde{J}_\parallel)$ and $B = D_\parallel/\tilde{J}_\parallel$. Here, we stress that the numerical value of $D_\parallel/k_B = 1.44$ K cannot serve as an estimate for the overall magnitude of the magnetic anisotropy which should be estimated from the band splitting of 50 $\mu$eV, i.e. $\approx 0.6$ K in units of Kelvin.

FIG. 5. Position of the triplet bands extracted from the neutron scattering data of Fig. 2. The dispersion drawn as a solid line represents a model with anisotropic interactions on the ladder legs (case d) as described in the text.

D. Final remarks

In BPCB the interactions on the ladder rungs are the strongest ones by far ($J_\perp \approx 3.6 J_\parallel$). On these bonds, antisymmetric exchange anisotropy is prohibited by symmetry and only Ising-type exchange anisotropy is allowed. It may thus seem plausible to expect this to be the dominant source of anisotropy. Yet, experimentally we find that the anisotropy is predominantly due to the much weaker leg interactions of the ladder. Indeed, this is fully consistent with the theory of super-exchange interactions, where symmetric and antisymmetric anisotropy contributions are always coupled. If either is prohibited by symmetry, also the other will vanish. The negligible Ising anisotropy on the ladder rungs in BPCB we thus see as supporting evidence, for the theory of anisotropic super-exchange\cite{5,6}.

Since the theory of super-exchange predicts DM
anisotropy $B \propto |D|$ and Ising anisotropy $A \propto |D|^2$, initially the latter was considered subdominant.\textsuperscript{22} However, both contributions are now understood to be equally important.\textsuperscript{23} Whilst not immediately intuitive, the triplet dispersions calculated in Sec.\textsuperscript{11} illustrate this effect: In the obtained expressions $A$ appears linearly, while $B$ only appears to second order as $B^2$.

Finally, we mention an apparent contradiction: Superexchange can never give rise to DM anisotropy exclusively. Yet, oftentimes pure DM interactions are used very successfully to explain anisotropy effects observed experimentally. Here, the present case study offers some illustration. For the spin ladder, different types of exchange anisotropy on the ladder legs lead to almost identical triplet dispersions that all explain our data equally well. We speculate that also in other systems one might encounter similar situations and it is for this reason that pure DM anisotropy is used so successfully to explain experimental findings, even though (within the theory of super-exchange) it lacks any microscopic justification. We are only aware of one experimental study on the helical triplet dispersions calculated in Sec.\textsuperscript{III C} illustrate this effect: In the obtained expressions $A$ appears exclusively. Yet, oftentimes pure DM interactions are used so successfully to explain experimental findings, even though (within the theory of anisotropic super-exchange) it lacks any microscopic justification. We are only aware of one experimental study on the helimagnet Ba$_2$CuGeO$_7$, where a model employing the full Hamiltonian for anisotropic super-exchange interactions (Eq.\textsuperscript{11}) was compared to experimental data and indeed better agreement was found than for a model employing DM anisotropy only.\textsuperscript{23}

Further case studies of the nature of magnetic exchange anisotropy would certainly be interesting. We also suggest, that whenever a model with pure DM interactions is considered, it would be enlightening to also consider a model with both Ising and DM anisotropy, compatible with super-exchange.

V. CONCLUSION

The prototypical spin ladder compound (C$_5$H$_{12}$N)$_2$CuBr$_4$ (BPCB) has been studied by means of high resolution inelastic neutron scattering. We find a small splitting of the triplet band of 50(1) $\mu$eV at the band minimum and 40(2) $\mu$eV at the band maximum. Further, for a spin ladder with exchange anisotropy, the triplet dispersion is calculated in a strong coupling expansion.

The Ising-type anisotropy allowed by crystallographic symmetry on the ladder rungs (by far the strongest bonds) we find to be negligible, in line with the theory of anisotropic super-exchange. Three models with exchange anisotropy on the ladder legs, all describe the data equally well. Whilst we cannot distinguish these by comparison to our data, we note that only one is compatible with super-exchange interactions.

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\textsuperscript{3} http://www.neutron.ethz.ch/
\textsuperscript{4} T. Moriya, Phys. Rev. Lett. 4, 228 (1960)
\textsuperscript{5} A. T. Savici, G. E. Granroth, C. L. Broholm, D. M. Pajerowski, C. M. Brown, D. R. Talham, M. W. Meisel, K. P. Schmidt, G. S. Uhrig, and S. E. Nagler, Phys. Rev. B 41, 1657 (1990)
\textsuperscript{6} A. A. Bazzi, V. K. Bhartiya, D. J. Voneshen, and A. Zhelev, Phys. Rev. Lett. 104, 094411 (2009)
\textsuperscript{7} C. Rüegg, K. Kiefer, B. Thielemann, D. F. McMorrow, V. Zapf, B. Normand, M. B. Zvonarev, P. Bouillot, C. Kollath, T. Giamarchi, S. Capponi, D. Poilblanc, D. Biner, and K. W. Krämer, Phys. Rev. Lett. 101, 247201 (2008)
\textsuperscript{8} M. Klanjšek, H. Mayaffre, C. Berthier, M. Horvatić, B. Chiari, O. Piovesana, P. Bouillot, C. Kollath, E. Orignac, R. Citro, and T. Giamarchi, Phys. Rev. Lett. 101, 137207 (2008)
\textsuperscript{9} B. Thielemann, C. Rüegg, H. M. Reznouk, A. M. Läuchli, J.-S. Caux, B. Normand, D. Biner, K. W. Krämer, H.-U. Gädel, J. Stahn, K. Habicht, K. Kiefer, M. Boehm, D. F. McMorrow, and J. Mesot, Phys. Rev. Lett. 102, 107204 (2009)
\textsuperscript{10} E. Czímár, M. Ozerov, J. Wosnitza, B. Thielemann, K. W. Krämer, C. Rüegg, O. Piovesana, M. Klanjšek, M. Horvatić, C. Berthier, and S. A. Zvyagin, Phys. Rev. B 82, 054431 (2010)
\textsuperscript{11} D. Bloser, V. K. Bhartiya, D. J. Voneshen, and A. Zhelev, Phys. Rev. Lett. 121, 247201 (2018)
\textsuperscript{12} The crystal structure plots shown in Fig. 1 are based on renderings generated by the VESTA software package.\textsuperscript{23}
\textsuperscript{13} I. Dzyaloshinsky, Journal of Physics and Chemistry of Solids 4, 241 (1958)
\textsuperscript{14} R. Bewley, J. Taylor, and S. Bennington., Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 637, 128 (2011)
\textsuperscript{15} The raw data is available in the STFC Research Data Repository.\textsuperscript{25}
\textsuperscript{16} M. Reigrotzki, H. Tsunetsugu, and T. M. Rice, Journal of Physics: Condensed Matter 6, 9235 (1994)
\textsuperscript{17} P. Fazekas, Lecture Notes on Electron Correlation and Magnetism (WORLD SCIENTIFIC, 1999)
\textsuperscript{18} W. Koshibae, Y. Ohta, and S. Maekawa, Phys. Rev. Lett. 41, 1657 (1990)
\textsuperscript{19} M. Reigrotzki, H. Tsunetsugu, and T. M. Rice, Journal of Physics: Condensed Matter 6, 9235 (1994)
\textsuperscript{20} P. Fazekas, Lecture Notes on Electron Correlation and Magnetism (WORLD SCIENTIFIC, 1999)
\textsuperscript{21} B. Thielemann, O. Entin-Wohlman, and A. Aharony, Phys. Rev. Lett. 71, 468 (1993)
\textsuperscript{22} In Fig. 4 the anisotropy parameter was chosen so as to ensure $\Delta_{\text{min}} = 50 \mu$eV. For the different models of leg anisotropy (Ising only, DM only, both), this gives
\( \Delta_{\text{max}} = 37, 32 \) and \( 35 \) \( \mu \)eV. However, since from \( 2^{\text{nd}} \) to \( 3^{\text{rd}} \) order perturbation expansion the dispersion still slightly changed, these small differences are probably not reliable enough to use as a criterion to select one of these three cases as the most suitable model describing the compound BPCB.

23 A. Zheludev, S. Maslov, I. Tsukada, I. Zaliznyak, L. P. Regnault, T. Masuda, K. Uchinokura, R. Erwin, and G. Shirane, Phys. Rev. Lett. 81, 5410 (1998).
24 K. Momma and F. Izumi, Journal of Applied Crystallography 44, 1272 (2011).
25 D. Blosser, V. K. Bhartiya, and A. Zheludev, RB1720009, STFC ISIS Facility (2018), 10.5286/ISIS.E.87813753.