Structure and magnetic properties of the $S = 3/2$ zigzag spin chain antiferromagnet BaCoTe$_2$O$_7$

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We report a study of the structure and magnetic properties of the $S = 3/2$ zigzag spin chain compound BaCoTe$_2$O$_7$. Neutron diffraction measurements show that it crystallizes in the noncentrosymmetric space group $Ama2$ with a canted $\uparrow\uparrow\downarrow\downarrow$ spin structure along the quasi-one-dimensional zigzag chain and a moment size of $1.89(2)$ $\mu_B$ at 2 K. Both magnetic susceptibility and specific heat measurements yield an antiferromagnetic phase transition at $T_N = 6.2$ K. A negative Curie-Weiss temperature, $\Theta_{CW} = -74.7(2)$ K, and an empirical frustration parameter, $f = |\Theta_{CW}|/T_N \approx 12$, are obtained by fitting the magnetic susceptibility, indicating antiferromagnetic interactions and strong magnetic frustration. From ultraviolet-visible absorption spectroscopy and first-principles calculations, an indirect band gap of $2.68(2)$ eV is determined. We propose that the canted zigzag spin chain of BaCoTe$_2$O$_7$ may produce a change in the polarization via the exchange-striction mechanism.

antiferromagnet, zigzag spin chain, magnetic frustration, spin structure

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1 Introduction

Quasi-one-dimensional (1D) systems have been extensively studied due to their intriguing properties [1-5].

Although an ideal 1D system does not become magnetically ordered at finite temperature [6], interchain interactions are inevitable in a real 1D material, which can result in a 3D magnetically ordered ground state [5].

Moreover, magnetic anisotropy arising from spin orbital coupling and crystal field environment constitute additional degrees of freedom that influence the magnetic order of a 1D system. In a spin-3/2 Co$^{2+}$ based 1D system, rich phase diagrams and certain emergent phenomena appear in a magnetic field or at high pressure owing to its strong magnetic anisotropy. Complicated magnetic field induced phase diagrams were observed in the Co$^{2+}$ based Ising spin chain systems BaCo$_2$V$_2$O$_6$ [7, 8] and CoNb$_2$O$_6$ [9, 10]. A plateau was
observed in the magnetization of the spin chain systems α-CoV$_2$O$_4$ [11, 12] and Ca$_2$Co$_2$O$_6$ [13, 14]. Thus, for a Co$^{2+}$, based 1D system, a combination of intra-and inter-chain interactions and magnetic anisotropy can produce various magnetic properties. It is therefore interesting to explore the exotic physics underlying the magnetic behaviors of the Co$^{2+}$-based 1D systems.

Compounds with the chemical formula BaMTe$_2$O$_7$ ($M$=Cu, Zn, Mg) belong to 1D chain systems that crystallize in the noncentrosymmetric space group Amma2 (Space group No. 40) [15, 16]. Their structures comprise a zigzag-chain arrangement of the transition metal atoms $M$ within the crystal field environment of the complex MO$_5$, in which the MO$_5$ entities are not directly connected by O$^{2-}$, as shown in Figure 1(a). The compounds BaMTe$_2$O$_7$ ($M$ = Zn, Mg) are nonmagnetic [16]. Although the spin of Cu$^{2+}$ is 1/2 in BaCuTe$_2$O$_7$, no magnetic order has been detected down to 1.8 K [15]. The similar Co$^{2+}$ based zigzag spin chain compounds α-BaCo$_2$X$_2$O$_7$ ($X$=P, As) have been investigated extensively [17,18]. They exhibit antiferromagnetic (AFM) orders below $T_N$=11 and 10 K, respectively. The susceptibility of α-BaCo$_2$X$_2$O$_7$ exhibits a large magnetic anisotropy. Their magnetization exhibits a 1/3 plateau below $T_N$, which results from the incommensurate atomic shifts. Because of the fascinating magnetic properties of the Co$^{2+}$ based zigzag spin chain systems, it is interesting to examine the properties of BaCoTe$_2$O$_7$. To our knowledge, there are no previous reports of the synthesis and characterization of BaCoTe$_2$O$_7$.

In this study, magnetic susceptibility, magnetization, specific heat, neutron powder diffraction (NPD), and ultraviolet-visible (UV-vis) absorption spectroscopy measurements as well as first-principles density functional theory (DFT) calculations were performed to determine the properties of BaCoTe$_2$O$_7$. A canted ↑↓↓↓ spin structure is identified below $T_N$ = 6.2 K, with a propagation vector $k$ = (0.5, 0, 0). A negative Curie-Weiss temperature $\Theta_{CW}$ = -74.7(2) K is obtained by fitting the susceptibility, which indicates that AFM interactions dominate. Large anisotropy is revealed on in the susceptibility and magnetization measurements. An indirect bandgap is determined as of 2.68(2) eV. The ordered moment of Co$^{2+}$ may be reduced by covalency between the Co$^{2+}$, 3d and O$^{2-}$, 2p orbitals and magnetic frustration.

2 Experiments and calculations

Single-crystal samples of BaCoTe$_2$O$_7$ were grown in two steps. First, a polycrystalline sample was synthesized using the conventional solid-state reaction. The starting materials BaCO$_3$ (99.99%), Co$_3$O$_4$ (99.9%), and TeO$_2$ (99.99%) were mixed thoroughly in the stoichiometric ratio in an agate mortar. The mixed powders were then pressed to a pellet and put into an alumina crucible covered with a lid. The pellet was calcined at 650°C in air in a muffle furnace for 7 d with several intimate grindings. Second, single-crystal samples were grown using the flux method. NaCl and KCl mixed in a 1:1 molar ratio were used as fluxes. The BaCoTe$_2$O$_7$ powder samples and fluxes with a weight ratio 1:0.75 were placed in a 3K; with a propagation vector $\mathbf{k} = (0, 0.5, 0)$. A negative Curie-Weiss temperature $\Theta_{CW} = -74.7(2)$ K is obtained by fitting the susceptibility, which indicates that AFM interactions dominate. Large anisotropy is revealed on in the susceptibility and magnetization measurements. An indirect bandgap is determined as of 2.68(2) eV. The ordered moment of Co$^{2+}$ may be reduced by covalency between the Co$^{2+}$, 3d and O$^{2-}$, 2p orbitals and magnetic frustration.

Our NPD experiment was conducted on the general pur-
pose powder diffractometer (GPPD) installed at the China Spallation Neutron Source (CSNS) [19]. The powder samples of BaCoTe₂O₇ were filled in a cylindrical vanadium can and measured at 2 and 300 K. The Rietveld method was employed to refine the NPD patterns using the FullProf suit package [20]. DC susceptibility and specific heat data were collected on a commercial physical property measurement system (PPMS, Quantum Design). The direction of the single crystal was determined by an X-ray Laue diffractometer. UV-vis spectroscopy measurements were conducted on an Ocean Optics DH-2000-BAL spectrometer.

We performed DFT calculations using the projector augmented wave [21] method with the generalized gradient approximation (GGA) of Perdew-Burke-Ernzerhof [22] functional as implemented in the Vienna \textit{ab initio} simulation package [23,24]. The kinetic cutoff energy was set to 550 eV and \Gamma-centered Monkhorst-Pack meshes of $2 \times 2 \times 3$ was used for the Brillouin-zone integrations [25]. For comparison with experiment, the lattice constants and spin structure refined from NPD at 2 K were employed. The atomic positions were fully relaxed using a conjugated gradient algorithm until the remaining force on each atom was $< 0.01$ eV/ Å. Spin-orbit coupling was considered in all the calculations. Moreover, we calculated the free energies of the different magnetic configurations.

3 Results and discussion

Figure 2 shows the observed and calculated NPD patterns of BaCoTe₂O₇. The refined structures reproduce the NPD patterns at 300 and 2 K very well when magnetic reflections are considered. No impurity phase is found. Our refinement reveals that BaCoTe₂O₇ crystallizes in the space group $Ama2$, which is isostructural to BaMTe₂O₇ ($M$=Cu, Zn, Mg). Table 1 lists the structural parameters obtained at 2 K. The composition from the NPD refinement is BaCo₀.₉₇Te₁.₉₀O₆.₆₄, close to the stoichiometric content. The refined parameters at 300 K are presented in the Supporting Information. As shown in Figure 1, each magnetic Co\textsuperscript{2+} ion is surrounded by five O\textsuperscript{2−} anions, thus forming a CoO$_{5}$ square pyramidal crystal field environment. The Te atoms exist in two valence states, Te(1)$^{6+}$ and Te(2)$^{4+}$, which yield the Te(1)O₆ and Te(2)O₄ crystal-field environments, respectively. The CoO$_{5}$, Te(1)O₆, and Te(2)O₄ entities are lined up in a zigzag chain running along the \textit{a} direction. Along the \textit{b} direction, Ba\textsuperscript{2+} ions separate the two dimensional [CoTe₂O₇]$^{2−}$ layers. In the \textit{c} direction, three zigzag chains stack as a periodic sequence of CoO₅, Te(1)O₆, and Te(2)O₄. This crystal structure was then confirmed by a single crystal XRD experiment (see the Supporting Information).

By comparing NPD patterns at 300 and 2 K, reflections from magnetic scattering can be clearly identified in Figure 2(b). All these magnetic peaks can be indexed with a propagation vector $\mathbf{k} = (0.5, 0, 0)$. A representational analysis was performed to determine the spin structure quantitatively using Basleps in the \textit{FullProf} suite package [26,27]. This results in the non-zero irreducible representation (IR) $\Gamma_1$. One Co\textsuperscript{2+}-ion crystallographic site is splitten into two sites Co\textsuperscript{2+}(1) $(x, y, z)$ and Co\textsuperscript{2+}(2) $(-x+1, -y+1/2, z-1/2)$. Table 2 lists the basis vectors (BV) for the two Co\textsuperscript{2+} sites of the IR $\Gamma_1$. The refinement with the IR $\Gamma_1$ reproduces the magnetic peaks at 2 K well, with the magnetic $R$ factor $R_{\text{mag}} = 4.27\%$. Consequently, the components of the magnetic moment along the \textit{a}, \textit{b}, and \textit{c} directions are $m_a = 0.0(1)$, $m_b = 0.60(4)$, and $m_c =$...
Table 1  Atoms, Wyckoff sites, fractional atomic coordinates (x/a, y/b, z/c), and occupations of BaCoTe$_2$O$_7$ at 2 K. The numbers in the brackets are the corresponding errors. The refined lattice constants are $a = 5.5685(1)$, $b = 15.1449(4)$, and $c = 7.2750(2)$ Å and the profile parameters are $R_p = 4.83\%$, $R_w = 4.68\%$, $R_e = 1.75\%$, and $\chi^2 = 7.19$.

| Atoms     | Wyckoff site | x/a   | y/b   | z/c   | Occupation |
|-----------|--------------|-------|-------|-------|------------|
| Ba        | 4b           | 1/4   | 0.2083(2) | 0.003(1) | 1.0        |
| Co        | 4b           | 1/4   | 0.1246(3) | 0.519(2) | 0.97(2)    |
| Te1       | 4b           | 1/4   | 0.9230(2) | 0.76(1)  | 0.95(2)    |
| Te2       | 4b           | 3/4   | 0.0719(2) | 0.250(1) | 0.95(2)    |
| O1        | 4b           | 3/4   | 0.1349(1) | 0.006(1) | 0.97(1)    |
| O2        | 8c           | 0.0028(8) | 0.1438(1) | 0.3115(9) | 1.98(3)    |
| O3        | 4b           | 1/4   | 0.9856(2) | 0.5357(8) | 0.93(2)    |
| O4        | 8c           | 0.0065(8) | 0.8428(1) | 0.6935(9) | 1.85(3)    |
| O5        | 4a           | 0     | 0     | 0.877(1) | 0.91(2)    |

Table 2  Basis vectors of the magnetic Co sites with the propagation vector $k = (0.5, 0.0)$. The atom sites of Co(1) and Co(2) are (x, y, z) and (−x + 1, −y + 1/2, z − 1/2), respectively.

| IR        | BV   | Co(1)       | Co(2)       |
|-----------|------|-------------|-------------|
| Γ$_1$     | $\phi_1$ | [1,0,0]    | [−1,0,0]    |
|           | $\phi_2$ | [0,1,0]    | [0,−1,0]    |
|           | $\phi_3$ | [0,0,1]    | [0,0,1]     |

1.79(2) $\mu_B$, thus yielding a total magnetic moment $m_{\text{tot}} = 1.89(2) \mu_B$. Figure 1(c) and (d) show different views of the spin structures so determined. The spins are arranged within the bc plane perpendicular to the zigzag-chain direction. Along the zigzag chain, adjacent spins are oriented at a relative angle of 37$^\circ$ and are arrayed in an alternating ferromagnetic (FM) and AFM way, which can be recognized as a canted $\uparrow\uparrow\uparrow\downarrow$ spin structure. Moreover, a similar magnetic structure, but with the only component of the magnetic moment oriented along the c-axis, is also tested; this yields $m = 1.86(1) \mu_B$ and $R_{\text{mag}} = 4.69\%$, which is worse than that for the canted spin structure. Figure 2(c) shows the NPD patterns for a magnetic peak at 2, 8, and 300 K. The broad peak at 8 K, which is above the Néel temperature, suggests the existence of magnetic frustration and short range magnetic correlations.

Figure 3(a) shows temperature dependence of the magnetic susceptibility $\chi$ measured under zero-field cooling (ZFC) and field cooling (FC) on a powder sample of BaCoTe$_2$O$_7$. No obvious difference is found between the ZFC and FC data. There is a broad peak around $T_{\text{max}} \approx 20$ K; this is a commonly observed characteristic for a 1D magnet because of the presence of magnetic correlations [5]. In the derivative $d\chi/dT$ shown in the inset of Figure 3(a), a peak appears at $T_N = 6.2$ K, which can be attributed to an AFM phase transition. Fitting the inverse susceptibility $1/(\chi - \chi_0)$ over the temperature range from 100 to 300 K to the Curie-Weiss law, $\chi = \chi_0 + C/(T - \Theta_{\text{CW}})$, yields the red solid line in Figure 3(a). The fitting shows that the contributions of core diamagnetism and Van Vleck paramagnetism $\chi_0$ amount to $-7.63(8) \times 10^{-4}$ emu Oe$^{-1}$ mol$^{-1}$, the Curie constant $C$ is $4.22(1)$ emu K mol$^{-1}$ Oe$^{-1}$, and the Curie-Weiss temperature $\Theta_{\text{CW}}$ is $-74.7(2)$ K. The negative value of $\Theta_{\text{CW}}$ demonstrates that AFM interactions dominate in the system. An empirical formula for the frustration parameter is $f = |\Theta_{\text{CW}}|/T_N = 12.08 > 10$, which suggests the possible existence of magnetic frustration or the effect of low dimensionality [28]. The effective magnetic moment is estimated to be $\mu_{\text{eff}} = 5.77(1) \mu_B$, which is larger than that of the theoretical value of $\mu_{\text{eff}} = g \sqrt{S(S+1)} = 3.87 \mu_B$ for a $S = 3/2$ system when assuming zero orbital residual and $g = 2$. The results signal a considerable orbital contribution from the Co$^{2+}$ ions, consistent with the reports for other Co$^{2+}$ based materials, such as BaCo$_2$X$_2$O$_7$ (X = P, As) [18], Na$_3$Co$_2$SbO$_6$ [29], BaCo$_2$(AsO$_4$)$_2$ [30].

Figure 3(b) and (c) present temperature dependence of the magnetic susceptibility $\chi$ and field dependent magnetization $M$ measured in the different crystallographic directions of a single crystal of BaCoTe$_2$O$_7$. The susceptibilities measured with the magnetic field applied along the $a$ direction ($\chi_a$), the $b$ direction ($\chi_b$), and the $c$ direction ($\chi_c$) exhibit a phase transition at $T_N \approx 6$ K, as seen in the derivative $d\chi/dT$, in agreement with that measured from the powder sample. In addition, these data exhibit large differences across the whole temperature range. For an isotropic 3D antiferromagnet, $\chi_a, \chi_b$, and $\chi_c$ should coincide in the high temperature range ($T > T_N$) [31]. The large anisotropy between $\chi_a, \chi_b$, and $\chi_c$ may be attributed to the anisotropy of the spin correlations persisting above $T_N$ and the 1D nature of BaCoTe$_2$O$_7$. For example, the Co$^{2+}$ based 1D compounds BaCo$_2$P$_2$O$_7$ [17], Ba$_2$CoSi$_2$O$_7$ [32], and BaCo$_2$V$_2$O$_8$ [7] also exhibit large anisotropies. The kinks in $\chi_a$ and $\chi_b$ at ~13 K in Figure 3 (b) may be related to impurities in the sample caused by contamination during the single-crystal growth process. The magnetization $M_c$ measured with a magnetic field applied along the $c$ direction exhibits a sharp change around
\[ \mu_0 H_c = 6 \text{T}, \] while there are no anomalies in \( M_s \) and \( M_b \). This is consistent with a spin flop-like transition for spins aligned in the \( bc \) plane with a dominant moment component \( m_v \).

Figure 4 shows temperature dependence of the specific heat \( C_p \) measured under zero and a finite magnetic field on a pellet pressed from a powder sample of \( \text{BaCoTe}_2\text{O}_7 \). A \( \lambda \)-like phase transition is observed at \( T_N = 6.2 \text{ K} \), consistent with the susceptibility measurements shown in Figure 3. To estimate the magnetic contribution \( C_{p,\text{mag}} \) on the specific heat, we subtracted the phonon part \( C_{p,\text{ph}} \) from the total \( C_p \); the electronic contribution is not considered because \( \text{BaCoTe}_2\text{O}_7 \) is an insulator. A modified Debye model is taken into account to estimate the phonon contribution, considering two phonon spectra: one being the spectrum of the light atoms (\( \text{O}^{2-} \) and the other the spectrum of the heavy atoms (\( \text{Ba}^{2+}, \text{Co}^{2+}, \text{Te}^{4+}, \) and \( \text{Te}^{6+} \)). This method was confirmed to be effective for analyzing \( \text{M}_2\text{MnTeO}_6 \) \( (M = \text{Sr}, \text{Ba}) [33, 34] \). The formula for the modified Debye model is

\[
C_{p,\text{ph}} = 9R \sum_{n=1}^{2} C_n \left( \frac{T}{\Theta_{Dn}} \right)^3 \int_0^{\Theta_{Dn}/T} \frac{x^4 \text{e}^x}{(\text{e}^x - 1)^2} \text{d}x. \tag{1}
\]

Fitting eq. (1) to the raw data over the temperature range from 60 to 200 K yields 6.5 light atoms and 4.5 heavy atoms out of the 11 atoms in a formula unit of \( \text{BaCoTe}_2\text{O}_7 \). The Debye temperatures, corresponding to the light and heavy atoms, are \( \Theta_{D1} = 855(26) \text{ K} \) and \( \Theta_{D2} = 256(7) \text{ K} \), respectively. This atomic ratio is in good agreement with the 7 light atoms (\( \text{O}^{2-} \) and 4 heavy atoms (\( \text{Ba}^{2+}, \text{Co}^{2+}, \text{Te}^{4+}, \) and \( \text{Te}^{6+} \)) in a formula unit of \( \text{BaCoTe}_2\text{O}_7 \). Based on the fitted parameters, the phonon part \( C_{p,\text{ph}} \) of the specific heat is extrapolated to 1.8 K, as shown by the black dashed line plotted in Figure 4.

Figure 3 (Color online) (a) Left axis: temperature dependence of the susceptibility \( \chi \) measured on a powder sample of \( \text{BaCoTe}_2\text{O}_7 \); the inset shows its derivative \( \chi'/DT \). Right axis: inverse susceptibility \( 1/(\chi - \chi_0) \) and the Curie-Weiss fit. (b) Temperature dependence of \( \chi \) measured along the three crystallographic directions of a single crystal; the inset shows their derivatives \( \chi'/DT \). (c) Magnetization measured in the three crystallographic directions of a single crystal; the inset shows an image of the single crystal with the crystallographic directions indicated.

Figure 4 (Color online) (a) Temperature dependence of the total specific heat \( C_p \), the phonon part \( C_{p,\text{ph}} \), and the magnetic part \( C_{p,\text{mag}} \) of \( \text{BaCoTe}_2\text{O}_7 \). The inset shows a magnified image of \( C_{p,\text{mag}} \) at low temperature. (b) Derived \( C_{p,\text{mag}}/T \) against temperature with the inset showing the magnetic entropy. (c) Specific heat \( C_p \) measured in different applied magnetic fields over the temperature range of 1.8-8 K.
have significant contributions in the energy range of $-5 \leq E \leq -1$ eV, indicating covalency between the 3d orbitals of Co$^{2+}$ and 2p orbitals of O$^{2-}$. The energy for the AFM $\uparrow\downarrow\downarrow\downarrow$ spin structure, $-5.8146$ eV/atom, is lower than that of the FM spin structure, $-5.7801$ eV/atom.

The calculated ordered moments are $m_a = 0$, $m_b = 0.89$, $m_c = 2.66$, and $m_{tot} = 2.80 \mu_B$, assuming the experimentally determined spin structure. The calculated ordered moment is smaller than the ideal local moment of $3 \mu_B$ for Co$^{2+}$ with a 3d$^7$ electronic configuration, which may be attributed to the covalency. But it is considerably larger than the experimental result. The Co$^{2+}$ ions in BaCoTe$_2$O$_7$ arrange in a triangular lattice in the zigzag chain and the competition between the intrachain exchange interactions $J_1 (d=4.658(8) \text{ Å at } T=2 \text{ K})$ and $J_2 (d=5.574(1) \text{ Å at } T=2 \text{ K})$ may lead to strong magnetic frustration, resulting in the reduced moment $1.89(2) \mu_B$ as observed in experiment. Note that weak interchain exchange interactions $J_3$ are necessary to stabilize the long range magnetic order.

The noncentrosymmetric BaCoTe$_2$O$_7$ is pyroelectric in its paramagnetic phase, but not ferroelectric. Because its polarization can not be reversed by an applied electric field [15, 16]. However, in the magnetically ordered phase, the spin structure with alternative FM and AFM arrangements may reduce the symmetry and result in a change in the electric polarization $\Delta P$ along the chain direction by inducing different exchange striction on the $\uparrow\uparrow$ and $\downarrow\downarrow$ spin pairs. $\Delta P$ may be partially reversible, which then presents a scenario similar to type II multiferroics [36-41]. Therefore, it is important to explore the magnetoelectric coupling in BaCoTe$_2$O$_7$, which we will leave for future work.

4 Conclusions

To summarize, we synthesized both powder and single-crystal samples of BaCoTe$_2$O$_7$ and investigated its structure, magnetic, and electronic properties. An antiferromagnetic $\uparrow\downarrow\downarrow\downarrow$ spin structure with a propagation vector $\mathbf{k} = (0.5, 0, 0)$ is revealed below $T_N = 6.2$ K. The large magnetic anisotropy at high temperatures suggests the existence of the magnetic exchange interactions above $T_N$. This field-induced behavior can be attributed to the effects of the single-ion anisotropy, the magnetic exchange interactions of Co$^{2+}$, as well as the 1D nature of this compound. Our DFT calculations suggest the existence of covalency between the Co$^{2+}$ 3d and O$^{2-}$ 2p orbitals. The covalency and magnetic frustration in the system may reduce its ordered moment. Further investigations on the spin dynamics and multiferroics in BaCoTe$_2$O$_7$ are necessary to reveal its unique magnetic and electric properties.

![Figure 5](image-url) (Color online) (a) Calculated total and partial DOS for BaCoTe$_2$O$_7$ and (b) the band structure based on the experimentally determined magnetic structure and $U = 6$ eV.
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Supporting Information

The supporting information is available online at phys.scichina.com and link.springer.com. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.

1. E. Ising, Z. Physik 31, 253 (1925).
2. W. Heisenberg, Z. Physik 49, 619 (1928).
3. H. Bethe, Z. Physik 71, 205 (1931).
4. I. Affleck, J. Phys.-Condens. Matter 1, 3047 (1989).
5. H.-J. Mikeska, and A. K. Kolezhuk, Lect. Notes Phys. 645, 1 (2004).
6. N. D. Mermin, and H. Wagner, Phys. Rev. Lett. 17, 1133 (1966).
7. Z. He, T. Taniyama, T. Kyômen, and M. Itoh, Phys. Rev. B 72, 172403 (2005).
8. B. Grenier, V. Simonet, B. Canals, P. Lejay, M. Klanjšek, M. Horvatić, and C. Berthier, Phys. Rev. B 92, 134416 (2015), arXiv: 1508.01815.
9. C. Heid, H. Weitzel, P. Burlet, M. Bonnet, W. Gonschorek, T. Vogt, J. Norwig, and H. Fuess, J. Magn. Magn. Mater. 151, 123 (1995).
10. C. Heid, H. Weitzel, P. Burlet, M. Winkelnmann, H. Ehrenberg, and H. Fuess, Phys. B-Condens. Matter 234-236, 574 (1997).
11. M. Lenertz, J. Alaria, D. Stoeffler, S. Colis, and A. Dinia, J. Phys. Chem. C 115, 17190 (2011).
12. C. B. Liu, Z. Z. He, S. L. Wang, M. Yang, Y. Liu, Y. J. Liu, R. Chen, H. P. Zhu, C. Dong, J. Z. Ke, Z. W. Ouyang, Z. C. XIA, and J. F. Wang, J. Phys.-Condens. Matter 31, 375802 (2019).
13. A. Maignan, C. Michel, A. C. Masset, C. Martin, and B. Raveau, Eur. Phys. J. B 15, 657 (2000).
14. S. Agrestini, L. C. Chapon, A. Daoud-Adaline, J. Schefner, A. Guikasov, C. Mazzioli, M. R. Lees, and O. A. Petrenko, Phys. Rev. Lett. 101, 097207 (2008), arXiv: 0807.4489.
15. J. Yeon, S. H. Kim, M. A. Hayward, and P. S. Halasyamani, Inorg. Chem. 50, 8663 (2011).
16. J. Yeon, S. H. Kim, S. D. Nguyen, H. Lee, and P. S. Halasyamani, Inorg. Chem. 51, 2662 (2012).
17. A. A. Belik, M. Azuma, and M. Takano, Inorg. Chem. 44, 7523 (2005).
18. R. David, H. Kabbour, S. Colis, A. Pautrat, E. Suard, and O. Mentré, J. Phys. Chem. C 117, 18190 (2013).
19. J. Chen, L. Kang, H. Lu, P. Luo, F. Wang, and L. He, Phys. B 551, 370 (2018).
20. H. M. Rietveld, J. Appl. Cryst. 2, 65 (1969).
21. P. E. Blöchl, Phys. Rev. B 50, 17953 (1994).
22. J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. 77, 3865 (1996).
23. G. Kresse, and J. Hafner, Phys. Rev. B 47, 558 (1993).
24. G. Kresse, and J. Furthmüller, Phys. Rev. B 54, 11169 (1996).
25. H. J. Monkhorst, and J. D. Pack, Phys. Rev. B 13, 5188 (1976).
26. E. F. Bertaut, J. Appl. Phys. Suppl. 33, 1138 (1962).
27. E. F. Bertaut, J. Phys. Colloques 32, C1-462 (1971).
28. L. J. de Jongh, and A. R. Miedema, Adv. Phys. 23, 1 (1974).
29. J. Q. Yan, S. Okamoto, Y. Wu, Q. Zheng, H. D. Zhou, H. B. Cao, and M. A. McGuire, Phys. Rev. Mater. 3, 074405 (2019), arXiv: 1905.09365.
30. R. Zhong, T. Gao, N. P. Ong, and R. J. Cava, Sci. Adv. 6, eaay6953 (2020), arXiv: 1910.08577.
31. R. L. Carlin, Magnetochemistry (Springer-Verlag, Berlin, 1986).
32. M. Soda, T. Hongo, M. Avdeev, H. Yoshizawa, T. Masuda, and H. Kawanou-Furukawa, Phys. Rev. B 100, 144410 (2019).
33. L. Ortega-San Martin, J. P. Chapman, L. Lezama, J. Sánchez Marcos, J. Rodríguez-Fernández, M. I. Arriortua, and T. Rojo, Eur. J. Inorg. Chem. 1362 (2006).
34. L. Li, N. Narayanan, S. Jin, J. Yu, Z. Liu, H. Sun, C. W. Wang, V. K. Peterson, Y. Liu, S. Danilkin, D. X. Yao, D. Yu, and M. Wang, Phys. Rev. B 102, 094413 (2020), arXiv: 2006.03356.
35. A. A. Aczel, L. Li, V. O. Garlea, J. Q. Yan, F. Weickert, M. Jaime, B. Maiorov, R. Movshovich, L. Civale, V. Keppens, and D. Mandrus, Phys. Rev. B 90, 134403 (2014), arXiv: 1407.4098.
36. B. Lorenz, Y. Q. Wang, and C. W. Chu, Phys. Rev. B 76, 104405 (2007), cond-mat/0608195.
37. Y. J. Choi, H. T. Yi, S. Lee, Q. Huang, V. Kiryukhin, and S. W. Cheong, Phys. Rev. Lett. 100, 047601 (2008).
38. R. Chen, J. F. Wang, Z. W. Ouyang, Z. Z. He, S. M. Wang, L. Lin, J. M. Liu, C. L. Lu, Y. Liu, C. Dong, B. C. Liu, Z. C. XIA, A. Matsuo, Y. Kohama, and K. Kindo, Phys. Rev. B 98, 184404 (2018).
39. W. H. Ji, L. Yin, W. M. Zhu, C. M. N. Kumar, C. Li, H. F. Li, W. T. Jin, S. Nandi, X. Su, T. Brückel, Y. Lee, B. N. Harmon, L. Ke, Z. W. Ouyang, and Y. Xiao, Phys. Rev. B 100, 134420 (2019), arXiv: 1905.06282.
40. S. Dong, J. M. Liu, and E. Dagotto, Phys. Rev. Lett. 113, 187204 (2014), arXiv: 1410.2682.
41. K. Du, L. Guo, J. Peng, X. Chen, Z. N. Zhou, Y. Zhang, T. Zheng, Y. P. Liang, J. P. Lu, Z. H. Ni, S. S. Wang, G. Van Tendeloo, Z. Zhang, S. Dong, and H. Tian, npj Quantum Mater. 5, 49 (2020), arXiv: 2007.12828.