Abstract

We report the magnetic properties of compounds in the KBaRE(BO$_3$)$_2$ family (RE = Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu), materials with a planar triangular lattice composed of rare earth ions. The samples were analyzed by x-ray diffraction and crystallize in the space group $R\overline{3}m$. Physical property measurements indicate the compounds display predominantly antiferromagnetic interactions between spins without any signs of magnetic ordering above 1.8 K. The ideal 2D rare earth triangular layers in this structure type make it a potential model system for investigating magnetic frustration in rare-earth-based materials.

Introduction

Geometrically frustrated magnetic materials are a fertile area of study in condensed matter physics. Competing interactions can lead to highly degenerate low temperature states and unconventional quantum phenomena, providing models for investigating exotic quasi-particle excitations and statistical mechanics. Built from corner-sharing magnetic tetrahedra, the 3D pyrochlore is a prime example of geometric frustration. These materials can exhibit a variety of magnetic properties, including spin ice (Dy$_2$Ti$_2$O$_7$, Ho$_2$Ti$_2$O$_7$) [1, 2], spin liquid (Tb$_2$Ti$_2$O$_7$) [3], long range ordered (Gd$_2$Ti$_2$O$_7$, GdSn$_2$O$_7$, Er$_2$Ti$_2$O$_7$) [4], and spin glass (Tb$_2$Mo$_2$O$_7$) behavior [5]. Recently, new compounds containing an ideal 2D rare earth (RE) kagome lattice, a subset of the pyrochlore lattice, have been reported (RE$_3$Sb$_3$Mg$_2$O$_{14}$ and RE$_3$Sb$_3$Zn$_2$O$_{14}$) [6, 7]. An interplay of the rare-earth-ion-dependent crystal field anisotropy and the frustrating lattice geometry generates their exotic behavior [8–10].

Perhaps the conceptually simplest example of magnetic frustration, however, is the 2D triangular lattice. The antiferromagnetic Ising model on a 2D triangular lattice was the first spin model realized to possess a disordered ground state and finite entropy at $T = 0$ K [11, 12]. Although the classical Heisenberg model on the triangular lattice does instead theoretically order at zero temperature, into a 120° three-sublattice pattern, the destiny of the quantum spin-$1/2$ model had eluded scientists for decades. Ultimately, it was determined that the quantum spin-$1/2$ model also yields ordering in the 120° three-sublattice pattern [13–15]. While this was not the resonating valence bond liquid state originally proposed for this kind of system [16, 17], that state was later found to be stable within a triangular lattice quantum dimer model [18]. It is thus evident that a simple generalization of triangular lattice magnetic models cannot be all-encompassing and that more studies are required to elucidate the physics behind these materials [19].

Extensive theoretical and experimental research has therefore been dedicated to the search for novel 2D frustrated materials to help advance the field. Recent studies have included those on the quantum spin liquid candidate YbMgGaO$_4$, which contains 2D triangular layers of Yb$^{3+}$ [20, 21]. Indeed, several models have been proposed where strong spin orbit coupling can introduce non-Heisenberg exchanges that effectively work to stabilize exotic quantum states such as the quantum spin liquid [21]. Because materials containing RE elements are natural candidates for finding strong correlation physics in the presence of strong spin orbit coupling, new materials of this type may lead to interesting magnetic ground states [22].

Here we present the elementary magnetic properties and lattice parameters of KBaRE(BO$_3$)$_2$ (RE = Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu), a series of materials with a 2D triangular lattice of RE ions. This structure type has previously been reported for RE = Tb, Y, Lu, Gd [23–26] The materials reported here are isostructural and crystallize with the Buetschliite [K$_2$Ca(CO$_3$)$_2$] mineral structure type in the space group $R\overline{3}m$. The distinct triangular layers constructed by RE$^{3+}$ in KBaRE(BO$_3$)$_2$ make this structure type a model system for exploring magnetic frustration on an ideal triangular lattice.
Experimental

The samples were synthesized by solid state reaction using the RE oxides, BaCO$_3$, K$_2$CO$_3$, and H$_3$BO$_3$ as starting materials. The RE oxides (Sm$_2$O$_3$, Eu$_2$O$_3$, Gd$_2$O$_3$, Tb$_2$O$_3$, Dy$_2$O$_3$, Ho$_2$O$_3$, Er$_2$O$_3$, Tm$_2$O$_3$, Yb$_2$O$_3$, and Lu$_2$O$_3$) were dried at 800 °C for one week prior to use. The carbonates were dried at 120 °C for several days. Stoichiometric mixtures (with 5% excess H$_3$BO$_3$ and 3% excess K$_2$CO$_3$ and BaCO$_3$) were ground thoroughly in an agate mortar and pestle, placed in alumina crucibles, and pre-reacted in air at 500 °C for 15 h. Following this, the samples were reground, reacted in air at 900 °C for 20 h, and then furnace-cooled to room temperature. KBaTb(BO$_3$)$_2$ was heated under flowing argon at 900 °C for an additional 10 h. The x-ray powder diffraction data were collected at room temperature using a Bruker D8 Advance Eco diffractometer with Cu Kα radiation (λ = 1.5418 Å) and a Lynxeye detector. Le Bail fits on KBaRE(BO$_3$)$_2$ were performed with the program Fullprof. A Quantum Design physical properties measurement system (PPMS) Dynacool was used to measure the DC magnetic susceptibilities for all samples between 1.8 and 300 K in a 5000 Oe applied field. In all samples, the magnetization was proportional to the magnetic field for temperatures exceeding 1.8 K and fields of up to 15 000 Oe. Therefore, the susceptibility was designated as $M/H$, where $H = 5000$ Oe.

Results and discussion

Crystal structure of KBaRE(BO$_3$)$_2$

The crystal structure of KBaRE(BO$_3$)$_2$ has been previously reported and is shown in figures 1 and 2 [23, 24, 26]. The materials reported here are all isostructural and crystallize in the space group R-3m. Several Le Bail profile fits to powder diffraction data from our samples are shown in figures 3 and 4, while the lattice parameters are provided in table 1. In the structure of KBaRE(BO$_3$)$_2$ (figure 1), layers of flat BO$_3$ triangular units extend orthogonal to the crystallographic c axis. Interlinked between these layers are double sheets of Ba/K atoms and single sheets of RE atoms. The RE layers are fully structurally ordered. Each 3$^+$ RE ion is octahedrally coordinated to oxygen, while the Ba$^{2+}$ and K$^+$ ions are mixed on the same site in a 9-coordinated Ba/K–O polyhedron. As emphasized in figure 2, the RE ions make up discrete triangular layers in the structure. The lattice parameters of the title compounds agree well with those previously reported [23–26]. As expected, the unit cell parameters decrease with decreasing ionic radius of the RE, as shown in figure 5 [27].

Magnetic properties

The magnetic data for KBaRE(BO$_3$)$_2$ was fit to the Curie–Weiss law $\chi = \frac{C}{T - \theta_W}$ where $\chi$ is the magnetic susceptibility, $C$ is the Curie Constant, and $\theta_W$ is the Weiss temperature. The effective moments were then obtained using the following relationship: $\mu_{\text{eff}} \propto 2.83\sqrt{C}$. The parameters for the fits are presented in table 2, along with comparisons to the expected moments for each RE$^{3+}$ ion [28]. At high temperatures, crystal field...
Figure 2. Schematic of KBaRE(BO₃)₂ structure displaying the positions of the metal ions in the unit cell. To the right of the schematic is an extended lattice showing the discrete RE³⁺ triangular planes in the structure.

Figure 3. Le Bail profile fits for KBaEu(BO₃)₂ (left) and KBaDy(BO₃)₂ (right). The experimental pattern is in red, the calculated pattern in black, and the difference plot in blue. The green marks indicate Bragg reflections.

Figure 4. Le Bail profile fits for KBaHo(BO₃)₂ (left) and KBaTm(BO₃)₂ (right). The experimental pattern is in red, the calculated pattern in black, and the difference plot in blue. The green marks indicate Bragg reflections.
effects may influence magnetic correlations and thereby impact susceptibility measurements; therefore, data was fit to the Curie–Weiss law at both low and high temperatures for comparison. Figures 6–14 feature Curie–Weiss fits of the samples reported here, along with field-dependent magnetization curves for the compounds at 2 K.

**KBaSm(BO3)2**

The high temperature susceptibility of KBaSm(BO3)2 in figure 6 reveals a large temperature independent Van Vleck paramagnetism contribution. This is characteristic of many Sm-containing compounds and arises from the first excited \( J = 7/2 \) multiplet of Sm\(^{3+} \) [29]. Therefore, the Curie–Weiss law was fit at the low temperature regime between 1.8 and 10 K, yielding a moment of 0.44 \( \mu_B/\text{Sm} \) and a Weiss temperature of \(-1.35 \text{ K}\). The negative Weiss temperature indicates the presence of antiferromagnetic nearest neighbor spin–spin interactions. While the \( \mu_{\text{eff}} \) is somewhat smaller than the expected free ion value of 0.83 \( \mu_B/\text{Sm} \), it is similar to that of other reported Sm\(^{3+} \)-containing oxides, such as Sm\(_3\)Sb\(_3\)Mg\(_2\)O\(_{14}\) (0.53 \( \mu_B/\text{Sm} \)) and Sm\(_2\)Zr\(_2\)O\(_7\) (0.50 \( \mu_B/\text{Sm} \)) [6, 30]. An effective moment as low as 0.15 \( \mu_B \) has been reported for Sm\(_2\)Ti\(_2\)O\(_7\) [30]. The low moments observed for these compounds are likely related to samarium’s large crystal field splitting of its lowest \( J = 5/2 \) multiplet. The field-dependent magnetization \( M(H) \) at 2 K in the left panel of figure 6 shows slight curvature but no indication of saturation up to an applied field of \( \mu_0 H = 9 \text{ T} \).

**KBaEu(BO3)2**

For Eu\(^{3+} \)-containing compounds, typically \( \mu_{\text{eff}} = 0 \). This arises because \( \mu_{\text{eff}} = g\sqrt{J(J + 1)} \), where for Eu\(^{3+} \), \( L = 3 \) and \( S = 3 \) and so \( J = 0 \). It is expected, therefore, that the ground state for Eu\(^{3+} \) may be non-magnetic [7], \( F_0 \). At low temperatures (up to roughly \(-100 \text{ K} \)), KBaEu(BO3)\(_2\)’s susceptibility displays Van Vleck paramagnetism (figure 7). This is characteristic of Eu\(^{3+} \) compounds, and is a result of sole population of the non-magnetic ground state. As the temperature rises, crystal field states originating from the first excited multiplet [7], \( F_1 \), are populated.

### Table 1. Lattice Parameters of KBaRE(BO3)\(_2\) determined from fits to room temperature powder x-ray diffraction data.

| RE     | RE ionic radius (Å) | \( a \) (Å)  | \( c \) (Å)  |
|--------|---------------------|----------------|----------------|
| Sm     | 0.958               | 5.491 12(4)    | 18.1138(7)     |
| Eu     | 0.947               | 5.482 99(9)    | 18.0410(4)     |
| Gd     | 0.938               | 5.475 36(4)    | 17.9790(3)     |
| Tb     | 0.923               | 5.463 42(6)    | 17.8893(3)     |
| Dy     | 0.912               | 5.455 36(2)    | 17.8360(8)     |
| Ho     | 0.901               | 5.448 27(3)    | 17.7647(2)     |
| Er     | 0.890               | 5.436 35(6)    | 17.7018(4)     |
| Tm     | 0.880               | 5.428 81(2)    | 17.6358(8)     |
| Yb     | 0.868               | 5.419 52(5)    | 17.6029(4)     |
| Lu     | 0.861               | 5.413 31(5)    | 17.5726(3)     |
This ultimately leads to a temperature-dependent contribution to KBaEu(BO₃)₂’s magnetic susceptibility. Although this effect may appear Curie–Weiss like, a fit of the data does not accurately reflect the exchange interactions, and so we do not apply the Curie–Weiss law to KBaEu(BO₃)₂. The field-dependent magnetization for KBaEu(BO₃)₂ at 2 K is shown in the right panel of figure 7. The \( M(H) \) plot is linear and reversible with the field increasing and decreasing.

**KBaGd(BO₃)₂**

The temperature dependent magnetic susceptibility of KBaGd(BO₃)₂ is shown in figure 8. As can be seen by both the susceptibility plot and the tabulated data in table 2, this material maintains strict Curie–Weiss law behavior to the lowest temperatures studied, with Weiss temperatures relatively small, on the order of 1 K. Indeed, fitting the

| RE  | High T \( p \) (high T) | \( \Theta_w \) (high T) | \( R^2 \) of fit | Low T \( p \) (low T) | \( \Theta_w \) (low T) | \( R^2 \) of fit | \( p_{exp} \) |
|-----|--------------------------|-------------------------|-----------------|--------------------------|-------------------------|-----------------|--------|
| Sm  | —                        | —                       | —               | 1.8–10                   | 0.43(9)                 | —               | 0.85   |
| Eu  | —                        | —                       | —               | 1.8–25                   | 7.70(1)                 | —               | 7.94   |
| Gd  | 150–300                  | 7.70(1)                 | 1.64(7)         | 0.99994                  | 1.8–25                  | 7.70(1)         | 0.99994 | 7.94   |
| Tb  | 150–300                  | 9.65(1)                 | —15.06(2)       | 0.99977                  | 10–25                   | 9.68(9)         | —11.09(7)| 9.72   |
| Dy  | 150–300                  | 10.62(5)                | —16.11(5)       | 0.99971                  | 15–30                   | 10.07(4)        | —5.41(7) | 10.63  |
| Ho  | 150–300                  | 10.50(1)                | —15.05(7)       | 0.99881                  | 10–25                   | 10.09(1)        | —3.09(9) | 10.60  |
| Er  | 150–300                  | 9.57(2)                 | —12.66(7)       | 0.99975                  | 8–23                    | 8.86(5)         | —3.09(5) | 9.59   |
| Tm  | 200–300                  | 7.58(3)                 | —29.37(1)       | 0.99915                  | 65–85                   | 7.80(5)         | —34.61(2)| 7.57   |
| Yb  | 125–275                  | 4.76(4)                 | —110.59(9)      | 0.99999                  | 1.8–25                  | 2.67(5)         | —0.83(7) | 4.53   |
| Lu  | —                        | —                       | —               | —                        | —                       | —               | —      | 0.0    |

The Curie–Weiss fit is shown in black. The inset is a magnified view of the low temperature inverse susceptibility. The right panel displays the field-dependent magnetization at 2 K.
high temperature data (150–300 K) yields a magnetic moment of 7.70 \(\mu_B/Gd\) and a Weiss temperature of 1.64 K. A low temperature fit (1.8–25 K) provides the same effective moment, but a negative Weiss temperature of \(-0.78\) K. The moment calculated here is close to the expected value of 7.94 \(\mu_B/Gd\) for the free ion \(\text{S}^{7/2}\) ground state of \(Gd^{3+}\). Similar moments with Weiss temperatures of a comparable magnitude were observed for the RE double perovskites Ba\(_2\)GdSbO\(_6\) (\(\mu_{\text{eff}} = 7.72; \Theta_w = -1.22\) K) and Sr\(_2\)GdSbO\(_6\) (\(\mu_{\text{eff}} = 7.64; \Theta_w = -0.14\) K) [31].

The \(M(H)\) plot at 2 K in the right panel of figure 8 reveals a nonlinear relationship between the magnetization and applied field and saturation at roughly 7.00 \(\mu_B/Gd\), slightly less than the moment calculated through the Curie–Weiss law and representative of all the Gd spins aligning in the powder sample in the direction of the applied field by roughly \(\mu_0H = 5\) T.

**KBaTb(BO\(_3\))\(_2\)**

The \(\chi(T)\) plot of KBaTb(BO\(_3\))\(_2\) is shown in figure 9. A high temperature fit of the data (150–300 K) gives an effective moment of 9.65 \(\mu_B/Tb\) and a Weiss temperature of -15.06 K. A similar \(\mu_{\text{eff}}\) of 9.69 and \(\Theta_w\) of \(-11.09\) were extrapolated from the low temperature regime. The moments calculated here are close to the expected free ion value of 9.72 \(\mu_B/Tb\). The antiferromagnetic Curie–Weiss temperature obtained from the high temperature fit is close to that observed for the pyrochlore Tb\(_2\)Ti\(_2\)O\(_7\) (\(\Theta_w = -17\) K) [32]. The 2 K field dependent magnetization plot of KBaTb(BO\(_3\))\(_2\) displayed in the right panel of figure 9 reveals a nonlinear response with some curvature but no signs of saturation up to an applied field of \(\mu_0H = 9\) T.

**KBaDy(BO\(_3\))\(_2\)**

Shown in figure 10 is the magnetic susceptibility and inverse susceptibility of KBaDy(BO\(_3\))\(_2\). A high temperature (150–300 K) Curie–Weiss fit of the data yields a moment of 10.63 \(\mu_B/Dy\) and a Weiss temperature of \(-16.12\) K, in agreement with the free ion moment of 10.63 \(\mu_B\) expected for Dy\(^{3+}\). The susceptibilities deviate toward smaller
values at the lowest temperatures measured, suggesting increasing antiferromagnetic correlations and perhaps emergent long-range order [31]. Indeed, fitting at low temperatures provide $\mu_{\text{eff}} = 10.07 \mu_B$/Dy and $\Theta_w = -5.42$. This low temperature moment is close to the value of $\mu_{\text{eff}} = 10.00 \mu_B$ for a ground state $mJ = \pm 15/2$ doublet, with $gJ = 4/3$ and corresponds well with that of Dy$_2$Ti$_2$O$_7$ [33]. The negative Weiss temperatures indicate antiferromagnetic interactions between spins. The field dependent magnetization at 2 K (figure 10, right panel) reveals nonlinear variation of the magnetization as a function of applied field and the onset of saturation at roughly $5 \mu_B$/Dy, about half the expected maximum of the effective moment. Saturation at about half the value of the effective moment is typically due to powder averaging of Ising spins.

**KBaHo(BO$_3$)$_2$**

Displayed in figure 11 is the $\chi(T)$ plot of KBaHo(BO$_3$)$_2$. The susceptibility behavior of the Dy analog is similar to that of KBaHo(BO$_3$)$_2$. A high temperature (150–300 K) fit of the inverse susceptibility results in a magnetic moment of 10.50 $\mu_B$/Ho and a Weiss temperature of $-15.05$ K. Applying the Curie–Weiss law to the low temperature regime yields the following parameters: $\mu_{\text{eff}} = 10.09 \mu_B$/Ho and $\Theta_w = -3.10$ K. The effective moments correspond well with the expected free ion value of 10.60 $\mu_B$/Ho and the value expected for an $mJ = \pm 8$ doublet ground state, $\mu_{\text{eff}} = 10.00 \mu_B$ [33]. The field dependent magnetization $M(H)$ plot at 2 K in the right panel of figure 11 shows curvature with the onset of saturation at about 5.5 $\mu_B$/Ho.

**KBaEr(BO$_3$)$_2$**

The magnetic susceptibility of KBaEr(BO$_3$)$_2$ is plotted in figure 12. Fitting the data from 150 to 300 K presents an effective moment of 9.57 $\mu_B$/Er and a Weiss temperature of $-12.67$ K, consistent with the free ion moment of $\mu_{\text{eff}} = 9.59 \mu_B$ for Er$^{3+}$ ($^4I_{15/2}$). A low temperature fit of the data yields $\mu_{\text{eff}} = 8.86 \mu_B$ and $\Theta_w = -3.09$ K.
in agreement with that of \( \mu_{\text{eff}} = 9.0 \mu_B \) for an \( m_J = \pm 15/2 \) doublet with \( g_J = 1.2 \) \([33]\). The field dependent magnetization at 2 K in the right panel of figure 12 shows a nonlinear response, approaching saturation at roughly 5.5 \( \mu_B/\text{Er} \), a bit more than half the free ion value.

**KBaTm(BO\(_3\))\(_2\)**

Exhibited in figure 13 is the magnetic susceptibility for KBaTm(BO\(_3\))\(_2\). Data were fit to the Curie—Weiss law in the temperature range 200–300 K, yielding an effective moment of 7.58 \( \mu_B \) and \( \Theta_w = -29.37 \text{ K} \). Fitting the data to a lower temperature range (65–85 K) provides \( \mu_{\text{eff}} = 7.80 \mu_B \) and \( \Theta_w = -34.61 \). The moments correspond well with that expected for Tm\(^{3+}\) (7.57 \( \mu_B \)) and the negative Weiss temperatures are suggestive of antiferromagnetic interactions between spins.

Due to crystal-field level splitting, Tm\(^{3+}\) (\( 4f_{12} \)) often takes on a singlet ground state. In a low symmetry site like that of Tm\(^{3+}\) in KBaTm(BO\(_3\))\(_2\), the \([3]H_2\) free ion state splits into a collection of single-ion crystal-field ground states where the strength and symmetry of the crystal field influence the energy separations \([34]\). As a result, the susceptibility features both a zero first order Zeeman contribution at low temperatures and a Van Vleck contribution due to the excited states. The anomaly in the \( \chi(T) \) plot for KBaTm(BO\(_3\))\(_2\) at around 15 K may therefore be interpreted as a Van Vleck component to the magnetic susceptibility. This anomaly was observed in all preparations of the Tm variant.

The \( M(H) \) plot at 2 K in figure 13 of KBaTm(BO\(_3\))\(_2\) reveals slight curvature without any signs of saturation up to an applied field of \( \mu_0H = 9 \text{ T} \).

**KBaYb(BO\(_3\))\(_2\)**

Plotted in figure 14 is the magnetic susceptibility of KBaYb(BO\(_3\))\(_2\). A high temperature fit of the data from 125–275 K gives an effective moment of 4.76 \( \mu_B/\text{Yb} \) and a Weiss temperature of \(-110.60 \text{ K} \). Fitting the data to
the Curie–Weiss law in the lower 1.8–25 K temperature regime yields $\mu_{\text{eff}} = 2.67 \mu_B$ and $\Theta_w = -0.84$ K. Similar temperature dependent behavior has been observed for Yb$_2$Ti$_2$O$_7$ [33, 35]. This trend is consistent with the free ion moment $\mu_{\text{eff}} = 4.54 \mu_B$ of Yb$^{3+}$ at room temperature and the movement of the ions into a Kramers doublet ground state at low temperature, with reduced moment. This behavior at low temperature is indicative of planar spin anisotropy. The field-dependent magnetization at 2 K in figure 14 sheds more light on this anisotropy and reveals a nonlinear magnetization response with the appearance of saturation at about 1.5 $\mu_B$/Yb.

**Conclusion**

We have synthesized and characterized several compounds in the KBaRE(BO$_3$)$_2$ family (RE = Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu). Le Bail profile fits were performed on the powder x-ray diffraction data from the compounds, from which the lattice parameters were extracted. They agree well with parameters previously reported in this structure type and show good evidence for the lanthanide contraction. Elementary magnetic property measurements show no signs of magnetic ordering above 1.8 K. Further low temperature characterization of these triangular RE materials, especially in single crystal form, is of significant interest; because many of these materials have been grown as single crystals [12–15], such studies are expected to be reasonably performed.

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**Figure 14.** Right panel: the magnetic susceptibility and reciprocal susceptibility of KBaYb(BO$_3$)$_2$ in an applied field of 5000 Oe. The Curie–Weiss fits are shown in black. Displayed in the left panel is the field-dependent magnetization at 2 K. The inset reveals the low temperature inverse susceptibility.
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