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Frustration and magnetism of the zigzag chain compounds \( \text{EuL}_2\text{O}_4 \) (\( L = \text{Yb, Lu, Gd, Eu} \))

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We present muon-spin rotation/relaxation and susceptibility measurements on polycrystalline samples of \( \text{EuL}_2\text{O}_4 \), where \( L \) is the lanthanide Yb, Lu, Gd, or Eu. The magnetic phase of these quasi-one-dimensional zigzag chain compounds is characterized with respect to the difference in their lanthanide magnetic moment. We find that the magnetic phase varies systematically with the lanthanide magnetic moment. At zero lanthanide moments (EuLu2O4), we find a static antiferromagnetic phase; as the moment increases, the phase gradually changes to an incommensurate spin-density-wave ordered phase, and finally reaches a dynamic phase, when large lanthanide magnetic moments are present (EuGd2O4).

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I. INTRODUCTION

One-dimensional systems that contain competing nearest-neighbor (NN) and next-nearest-neighbor (NNN) interactions offer an intriguing example of relatively simple systems which show fascinating magnetic phenomena and complex multiple phases. Over the last decade, the successful synthesis of one-dimensional (1D) zigzag chains has presented a remarkable diversity of magnetic phenomena, inspiring experimental1–5 and theoretical6,7 studies. Such theoretical studies have shown that the XXZ Hamiltonian, appropriate for quasi-1D systems, can be characterized by only two parameters. Interestingly, this model is shown to contain multiple phases: e.g., vector chiral orders, dimer orders, Néel order, and a Tomonaga-Luttinger liquid.8 This scenario has also been shown in the presence of an external field in the Heisenberg model.9,10 Thus, the question arises whether we can control the magnitude of the magnetic moments of the zigzag chain and use it as a tuning parameter to manipulate the magnetic frustration-ground state interplay in these systems. Here we attempt to answer this question by studying new compounds, in which the frustration is tuned by the chemical substitution of the magnetic ions.

The \( \text{EuL}_2\text{O}_4 \) compounds, where \( L \) is a lanthanide element, are composed of a \( L \) zigzag chains with a honeycomb-like structure formed by these chains, with the crystallographic CaFe2O4 (CFO)-type \( \text{Pnma} \) structure.11 The schematic structure is shown in the inset of Fig. 1 where possible exchange paths which create an \( ac \)-plane honeycomb lattice as well the zigzag along the \( b \) axis are suggested by the black lines. This crystallographic structure is the basis of numerous recently reported quasi-1D zigzag chains.12,13 Due to the high neutron absorption cross section by Eu, neutron scattering experiments on such compounds are challenging; thus, \( \mu \)SR has a key advantage in this case for providing information on the magnetic interactions. Here, we report on an extensive \( \mu \)SR study of EuL2O4, where \( L = \text{Eu, Gd, Yb, and Lu} \), in order to clarify their magnetic nature with respect to their frustration. The lanthanides Eu, Gd, Yb, and Lu are very similar in their atomic and electronic properties. Consequently, comparative studies on EuL2O4 allow a unique comparison of the magnetic interactions, since Yb, Gd, and Eu have different magnetic moments, while Lu has none. Hence, we are able to probe how the magnetic ground state is affected, from a situation where there are no magnetic zigzag chains, or no frustration along the zigzag ladders when there is no magnetic moment (in \( L = \text{Lu} \)), to a state where we have intense frustration with a large magnetic moment (as in \( L = \text{Gd} \)). Naturally, the increase in the magnetic moment can influence the ground state by embodying high-order perturbation such as NNN interactions, hence this study could potentially provide theorists an interesting challenge: to predict high-order perturbations and the resulting ground state due to the increase of the magnetic moment in this system.

Similar compounds, SrL2O4 (Ref. 14) and BaL2O4,15 were reported to show an anomaly in susceptibility measurements, which is ascribed to the magnetic interaction between the \( L^{3+} \) ions. This is because the alkali ions, Sr2+ and Ba2+, should not contribute to the magnetism due to their diamagnetic nature. By contrast, the Eu2+ ions at the alkali site in EuL2O4 are expected to introduce additional magnetic interactions with the \( L^{3+} \) ions, thus affecting the magnetic behavior, due to their large magnetic moment and partially occupied \( 4f \) orbital.

The manuscript is organized as follows: in Sec. II, we illustrate the EuL2O4 experimental details on the preparation and the magnetic characterization using bulk magnetization and \( \mu \)SR. In Sec. III, we present the results from the \( \mu \)SR measurements and describe the analysis following by a discussion.

II. EXPERIMENT

Polycrystalline EuL2O4 samples were prepared at Hokkaido University by a solid-state reaction between EuO
and $L_2O_3$ in an evacuated quartz tube at 1373 K for 24 h, as described in Ref. 11. Prior to this reaction, EuO was prepared by heating mixtures of Eu metal and Eu$_2$O$_3$ in a quartz tube at 1073 K for 24 h. dc-$\chi$ measurements and powder diffraction (Cu K$\alpha$ radiation) analyses for the present samples yielded reproducible results consistent with those in a previous report.

Bulk susceptibility ($\chi$) of Eu$L_2O_4$ were measured using a Quantum Design Magnetic Properties Measurement System superconducting quantum interference device (SQUID). These $\chi$ measurements showed that these Eu$L_2O_4$ compounds are very similar in their bulk magnetic behavior showing an antiferromagnetic (AF) transition at $T \approx 6$ K (for example, SQUID magnetometer measurements of $L = $ Eu, Lu are presented in Figs. 4, and for $L = $ Gd and Yb are shown in Figs. 4 and 5, respectively). Hence the 3D nature of the transition is mainly controlled by the Eu$^{2+}$ ion. Although bulk $\chi$ measurements on EuLu$_2$O$_4$ indicate the formation of AF order below $T_N = 5.7$ K, its positive Curie-Weiss temperature [$\theta_{CW} = 9.8(7)$ K] suggests a predominance ferromagnetic (FM) interaction of the magnetic Eu$^{2+}$ ions. The suggested resolution is a FM coupling between NN Eu$^{2+}$ ions together with AF interaction between parallel Eu$^{2+}$ 1D chains, realizing an overall AF structure.\textsuperscript{11} Note that the Lu$^{3+}$ ion’s $4f$ orbitals are fully occupied, resulting in the nonmagnetic nature of the Lu$^{3+}$ ions. A Curie law fitting of $\chi$ at 200 $\leq$ $T$ $\leq$ 400 K reveals the effective magnetic moment to be $\mu_{\text{eff}} = 7.94(2)\mu_B$ as expected from the Eu$^{2+}$ ions. This enables us to calculate the effective moment of the $L_2^3 = $ Gd, Yb, and Eu ion by removing the contribution of the Eu$^{2+}$ using $2\mu_l^2 = \mu_{\text{eff}}^2 - 7.94^2$ (see Table I), where $\mu_{\text{eff}}$ is the effective magnetic moment extracted from the $\chi$ measurements of the Eu$L_2O_4$. These $\mu_l$ values obtained are found to slightly deviate from the theoretically calculated ion.\textsuperscript{16} The magnetic interactions between the $L_2^3$ should therefore contribute to the magnetic behavior; as a result, the triangular frustrated geometry on the LO$_2$ zigzag chain should play a role in determining their magnetic ground states.

In the $\mu$SR experiments, the powder sample was placed in a small Al-coated Mylar envelope. The sample was then placed onto a low-background sample holder in a liquid-He flow cryostat on the M20 surface muon channel at TRIUMF, Vancouver, Canada. Zero-field (ZF) and weak-transverse-field (wTF) spectra were gathered in the $T$ range between 1.8 and 30 K. The ZF-$\mu$SR technique is a sensitive probe of local magnetic [dis]order through the precession of the muon spin in internal magnetic fields at the muon’s interstitial sites. In the wTF-$\mu$SR technique, a field is applied perpendicular to the muon-spin direction; the applied field $H_{\text{TF}}$ is weak compared to any spontaneous internal fields in a magnetic ordered phase. Thus, the amplitude (asymmetry) of the $\mu$SR precession at the frequency of $H_{\text{TF}}$ is a measure of the volume fraction of the sample that does not exhibit local magnetic order.\textsuperscript{17,18}

### III. Results

#### A. $L = $ Lu, Eu, Yb

To probe the magnetic phase of Eu$L_2O_4$, we concentrate mainly on the $\mu$SR spectra measured below $T_N$. The raw ZF data taken for $L = $ Lu, Eu, and Yb, is unlike the $L = $ Gd compound, hence is discussed subsequently. As a typical example of the spectra for $L = $ Lu, Eu, and Yb, Fig. 2(a) depicts the raw ZF data taken at $T = 1.8$ K. An oscillation due to the ordered state is observed and as an example the Fourier transform (FT) for $L = $ Lu is shown in Fig. 2(b). The spectrum clearly indicates a single frequency at $\sim$60 MHz. The ZF-$\mu$SR spectra can be fitted with the sum of an exponentially relaxing

<table>
<thead>
<tr>
<th>$L$</th>
<th>$T_N$ (K)</th>
<th>$\theta_{CW}$ (K)</th>
<th>$\mu_L$ ($\mu_B$)</th>
<th>$\varphi$ (deg.)</th>
<th>$F(t)$</th>
<th>$\chi^2$/$\text{NDF}$</th>
<th>$\beta_{\text{BCS}}$</th>
<th>$\beta(T &gt; T_N)$</th>
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</thead>
<tbody>
<tr>
<td>Lu</td>
<td>5.7</td>
<td>9.8(7)</td>
<td>...</td>
<td>1.67(17)</td>
<td>$\cos(\omega_{\text{ZF}} + \varphi)$</td>
<td>1.3(1)</td>
<td>0.489(3)</td>
<td>0.65(3)</td>
</tr>
<tr>
<td>Eu</td>
<td>5.4</td>
<td>$-32.7(6)$</td>
<td>3.83(1)</td>
<td>42(20)</td>
<td>$J_0(\omega_{\text{ZF}})$</td>
<td>1.03(5)</td>
<td>0.36(2)</td>
<td>0.58(3)</td>
</tr>
<tr>
<td>Yb</td>
<td>6.3</td>
<td>$-32.2$</td>
<td>4.77(1)</td>
<td>158(6)</td>
<td>$J_0(\omega_{\text{ZF}})$</td>
<td>1.08(7)</td>
<td>0.306(4)</td>
<td>0.68(4)</td>
</tr>
<tr>
<td>Gd</td>
<td>5.6</td>
<td>$-4.8(3)$</td>
<td>7.82(2)</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>Dynamic AF</td>
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TABLE I: Key results of the bulk and local data obtained by SQUID and $\mu$SR. $T_N$ and $\theta_{CW}$ are the Neél and Curie-Weiss temperatures, respectively, $\mu_L$ is the effective moment of the $L$ ion. $\varphi$ is the initial phase measured at $T = 1.8$ K with a cosine fitting function [Eq. (1)]. $F(t)$ is the fitting function as described in the text, $\chi^2$/$\text{NDF}$ is the reduced $\chi^2$ goodness of fit parameter when the raw data are fitted with $F(t)$. $\beta_{\text{BCS}}$ is the BCS energy gap critical exponent, the ground state is obtained from the ZF-$\mu$SR data below $T_N$, and $\beta$ is the critical exponent measured above $T_N$. 
However, due to the fast damping of the oscillating signal, a fast relaxing component and a 1/3 tail due to the powder form of the samples,

\[ A_0 P_{ZF}(t) = A_0 \left[ A_{\text{slow}} \left( 2 \cos(\omega_{AF}t + \psi) + \exp(-\lambda_{AF}t) \right) + \frac{1}{2} \exp(-\lambda_{tail}t) + A_{\text{fast}} \exp(-\lambda_{fast}t) \right], \]  

where \( A_0 \) is the initial asymmetry, determined by measurements of a Ag plate prior to the sample measurements. The frequency \( \omega_{AF} = 2\pi f \) and relaxation rate \( \lambda_{AF} \) arise from the ordered AF phase. The fast relaxing component is due to fluctuating moments, which give only a minor contribution, as evidenced by its small asymmetry, \( A_{\text{fast}} < A_0 \).

The oscillating signal in the spectra could also be fitted with a zeroth-order Bessel function of the first kind \[ J_0(\omega_{AF}t) \] instead of a cosine \( \cos(\omega_{AF}t + \psi) \) in Eq. (1). This has been found to be appropriate in numerous quasi-1D systems.\(^{3,18}\) A ZF spectrum described by \( J_0 \) is a key signature that the muon experiences an incommensurate (IC) field distribution, and as such points to a spin-density-wave (SDW) ground state.\(^{19}\) However, due to the fast damping of the oscillating signal, it is hard to justify one function over the other.\(^{20}\) One of the characteristics of an IC-SDW which suggests a Bessel function fit is a nonzero phase, \( \psi \). In EuEu\(_2\)O\(_4\) and EuYb\(_2\)O\(_4\), the phase changes with \( T \) particularly below the vicinity of \( T_N \) with \( \psi(T = 1.8 K) = 42(20)^\circ \) and 158(6)\(^\circ \) > 0, respectively. Hence, these compounds are more likely to possess a mixed IC-AF phase, or an IC phase. The real part of the FT, which reflects the local field distribution, also shows the characteristic tail of an IC modulation in these compounds [see inset in Fig. 2(b)]. In contrast, the EuLu\(_2\)O\(_4\) shows a negligible initial phase \( [\psi(T = 1.8 K) = 1.67(17) \neq 0^\circ] \); thus, it is more likely to assume a commensurate magnetic phase.

We can also use the wTF data as a complementary measurement. In the paramagnetic phase, the wTF-spectrum is fitted with a slow exponentially relaxing cosine function:

\[ A_0 P_{TF}(t) = A_{TF} \cos(\omega t + \psi) \exp(-\lambda t), \]  

where \( \omega = \gamma_{\mu} H_{TF} \) is the muon Larmor frequency, with \( \gamma_{\mu} = 13.554 \) kHz/G, and \( H_{TF} = 30 \) Oe is the applied wTF. Below \( T_N \), an additional term is needed to fit the signal, due to the AF ordered phase:

\[ A_0 P_{TF}(t) = A_{TF} \cos(\omega t + \psi) \exp(-\lambda_{AF} t) + A_{AF} F(t) \exp(-\lambda_{AF} t), \]  

where \( F \) is a cosine or a Bessel function, as in the ZF measurements. As an example, the raw wTF-spectra of \( L = Lu \) obtained at 1.7 and 8.0 K are plotted in Fig. 2(c).

Figure 3 shows the \( T \) dependence of \( f \) extracted from the fits for the \( L = Lu, Eu, \) and Yb samples. Obviously, the moment size is not the sole parameter which influence the internal field and therefore the muon precession frequency, for example, the magnetic order should also be taken into consideration. Nevertheless, the difference between the three compounds are apparent in the slopes of the \( f(T) \) curve, for instance, the slope below \( T = 4 \) K is larger in \( L = Lu \) than that of Eu and Yb. The common feature, however, is the behavior of \( f(T) \) which is typical of SDW order. That is, the \( f(T) \) curve follows the theoretical \( T \) dependence of the BCS gap energy,\(^{21}\) i.e., \( f = f_{BCS} \propto \tanh[1.74 \times (1 - T/T_N)\beta_{BCS}] \), as seen by the solid lines in Fig. 3. The parameter \( \beta_{BCS} \) depends on the Hamiltonian, or phase, describing the magnetic interaction in the experimental system. In EuLu\(_2\)O\(_4\), the fit to this equation yields the expected \( \beta_{BCS} = 1/2 \) from mean-field theory\(^{22}\) \[ [\beta_{BCS} = 0.489(3)] \]. Whereas in EuEu\(_2\)O\(_4\) and EuYb\(_2\)O\(_4\), it reveals \( \beta_{BCS} \) is decreasing with increasing magnetic moment \[ [\beta_{BCS} = 0.36(2) \text{ and } \beta_{BCS} = 0.302(4), \text{ respectively}] \]. As an example, \( \beta_{BCS} = 0.326 \) is expected in the one-spin-dimensionality Ising lattice.\(^{23}\) Regardless of the absolute value of \( \beta_{BCS} \), the difference between the values in the different compounds implies that the large Yb moments along the zigzag chain affect the magnetic order and alters the ground state.

\section*{B. \( L = Gd \)}

In EuGd\(_2\)O\(_4\), on the other hand, ZF and wTF measurements below \( T_N \) do not show a clear formation of static AF order. Instead a slow relaxation is observed, as seen in the inset of Fig. 4. This was also confirmed using the Hitime spectrometer on the M15 channel at TRIUMF, which currently has the highest time resolution in the world. The ZF spectrum is well fitted globally by a stretched exponential relaxation function:

\[ A_0 P_{ZF}(t) = A_0 \exp\left[-\left(\lambda_{ZF} t\right)^{\beta}\right], \]  

FIG. 2. (Color online) (a) The raw ZF data of EuLu\(_2\)O\(_4\), taken at \( T = 1.8 \) K, solid lines demonstrate the fit. The data of \( L = Eu, Yb \) are shifted by +0.1 and +0.2 for clarity. (b) The ZF FT of EuLu\(_2\)O\(_4\), taken at \( T = 1.8 \) K; inset shows the real part of the FT of EuEu\(_2\)O\(_4\) \( T = 1.8 \) K. (c) Measurements in wTF taken at 1.7 and 8.0 K; the asymmetry of the 1.7 K data is shifted by 0.05 for clarity. The solid lines indicate the fits.

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where $\beta = 0.42(8)$ and $A_0$ were common for all $T$. Here, $\beta = 1/2$ suggests a dilute disordered magnet,\textsuperscript{24} whereas $\beta = 1/3$ characterizes a dense disordered phase.\textsuperscript{25} As expected, the longitudinal relaxation ($\lambda_{ZF} = 1/T_1$) shows critical behavior as $T \to T_N$. The main panel of Fig. 4 displays the $T$ dependence of $\lambda_{ZF}$ along with the $\chi(T)$ curve of EuGd$_2$O$_4$. Clearly, the ZF-$\mu$SR data tracks the bulk $\chi$ measurement. Since the two measurements (local and bulk) do not indicate a Curie tail, which is expected at low $T$ due to impurities, the absence of static AF order, detected by ZF-$\mu$SR, below $T_N$ is most likely an intrinsic feature of EuGd$_2$O$_4$, and not due to impurities. Hence, we argue that the strong Gd moments destroy the local magnetic order, whereas such order is seen in the other compounds having smaller moments. This conclusion is supported by the fact that EuYb$_2$O$_4$, in which Yb has the second largest magnetic moments in our study, also shows a weak IC-SDW behavior. Hence, we claim that in $L = \text{Lu}$, the AF order is caused only due to the Eu$^{2+}$ ions ($7.94 \mu_B$), while in $L = \text{Eu}$ and Yb, the moment perturb the AF order of the Eu$^{3+}$. In $L = \text{Gd}$, the moment is comparable to the Eu, resulting in a small internal field at the muon site. Note that

\[ \chi(T) = \frac{C}{T - \Delta^2} \]

where $C = 0.16(2)$ was common for all $L$. Here, $\chi(T)$ was fitted with a phenomenological stretched exponential [as in Eq. (4)], with a faster relaxing component, as indicated by the fits. The initial asymmetry $A_0$ and $\beta = 0.68(4)$ were common in all data sets. On the right axis of Fig. 5(a), we plot the ZF-$\mu$SR longitudinal relaxation rates, $T_{1}^{-1}$, extracted from the fits. The $T_{1}^{-1}$ data clearly fits the $\chi(T)$ data, showing the enhancement and slowing down of the fluctuations as $T \to T_N$. Such measurements were also performed on the paramagnetic phases of EuLu$_2$O$_4$ and EuEu$_2$O$_4$. Spin-fluctuation theories suggest that $T_{1}^{-1} \propto \chi$.\textsuperscript{26} Thus, the main panel of Fig. 5(b) shows plots of $(T_{1}T)^{-1}$ versus $\chi$ for these compounds. Fits to this function, indicated by the solid lines, reveal $n = 1.15(5), 1.25(6)$, and 0.72(2), for $L = \text{Lu}$, Eu, and Yb, respectively. The negative curvature of EuYb$_2$O$_4$ suggests a slightly different scenario, where the relaxation is driven by random field fluctuations,\textsuperscript{27} hence $(T_{1}T\chi)^{-1} = \Delta T/(1 + \omega^2 T^2)$, where $\omega \propto T^{-n}$ is the
correlation time and \( \omega \) is the Larmor frequency at the muon site. We find \( \tau = T^{-0.684(2)} \). This fit is represented in the inset of Fig. 5(b).

To summarize, here we were able to show that with the synthesis of different zigzag magnetic moments, we can tune the system into a specific ground state phase. \( \mu SR \) and susceptibility measurements were performed on four Eu\( _2 \)O\( _3 \) compounds (where \( L \) is the lanthanide, Eu, Gd, Yb, and Lu). The bulk \( \chi \) data reveal that an AF transition takes place in all at \( 5.4 \leq T \leq 6.3 \) K. On a local scale, \( \mu SR \) indicates the formation of a static AF (in \( L = \text{Lu} \), a mixed AF/IC-SDW (\( L = \text{Eu} \)), and an IC-SDW (\( L = \text{Yb} \)) ordered phase. In EuGd\( _2 \)O\( _4 \), a dynamic disordered phase is suggested. Thus, the difference in the magnetic moment, arising from the lanthanide ion (\( L^{3+} \)), in these quasi-1D zigzag chains reveals that static AF order becomes dynamic as the magnetic moment along the zigzag chain increases.

\[ \text{ACKNOWLEDGMENTS} \]

We are grateful to the staff of TRIUMF for assistance with the \( \mu SR \) experiments. J.H.B. is supported at UBC by NSERC of Canada and (through TRIUMF) by NRC of Canada. K.H.C. is supported by NSERC of Canada and (through TRIUMF) by NRC of Canada.

\[ \text{In EuLu}_2\text{O}_4, the } T - \text{averaged reduced- } \chi^2 \text{ value achieved from the fit to Eq. (1) was } 1.30(18) \text{ and } 1.31(15) \text{ from the fit with Eq. (1)} \]

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