I. INTRODUCTION

Geometrical frustration arises in systems where the interactions between the magnetic moments are incompatible with their spatial arrangement in a lattice, so that at low temperatures not all of the interaction energies can be simultaneously minimized [1–3]. Thus, in the presence of antiferromagnetic exchange interactions, many magnets based on corner- or edge-sharing triangles [4–6] or tetrahedra [7–9] can exhibit geometric frustration. A general consequence of frustration is the establishment of magnetic order at temperatures much lower than what would be expected from the strength of the exchange interactions, or even the absence of magnetic order down to the lowest measured temperatures [10–13]. While some frustrated systems remain in a highly degenerate manifold of ground states, others form a unique ground state at very low temperatures. In this regime, small perturbations to the Hamiltonian, such as further-neighbor exchange, single-ion anisotropy, magnetic dipolar interactions, etc., as well as other effects such as quantum fluctuations, become important and can lead to unusual spin arrangements. New geometrically frustrated systems are constantly being discovered, and their behavior is often complex, with a rich variety of low-temperature properties.

The SrL2O4 (L = Lanthanide) family of compounds [14], which crystallize in the form of calcium ferrite [15], space group Pnam, have been suggested as lattices that could give rise to frustrated magnetism. For the case of SrGd2O4, the magnetic Gd3+ ions are linked in a network of triangles and hexagons, as shown in Fig. 1, and there are two crystallographically inequivalent sites for the rare-earth ions (which are shown in red and blue). Along the c axis, the magnetic ions form zigzag chains and the nearest distance between the Gd3+ ions is about 3.48 Å [16]. The zigzag chains (shown with either red or blue bonds), which consist of the Gd3+ ions sitting in the same crystallographic positions have shorter Gd3+-Gd3+ separations across the zigzags (3.54 to 3.62 Å) as compared to the longer bonds (shown in gray, 3.85 to 4.12 Å), which connect the Gd3+ ions at inequivalent positions [16]. The zigzag structures can be frustrated if the dominant exchange is antiferromagnetic, and are magnetically equivalent to spin chains with first- and second-nearest-neighbor interactions. The chains of Gd3+ ions interconnect by forming a distorted honeycomb structure, a bipartite lattice made up of edge sharing hexagons, in the a-b plane.

The magnetic characterization of powder samples of the SrL2O4 family of compounds began with the work of Karunadasa et al. [17]. In this early study, measurements of the magnetic susceptibility for all of the SrL2O4 compounds had revealed a disparity in the measured Weiss temperatures, ΘCW, and the lack of long-range order down to 1.8 K [17]. Single crystals of these oxides have been grown [18,19], and four members of the SrL2O4 family, where L = Dy [20], Ho [21–25], Er [21,26], and Yb [19] have been studied in detail. Despite the large reported values of the Weiss temperatures (−13.5 to −99.4 K) [17], measurements of the low-temperature susceptibility and heat capacity on the SrHo2O4 [21], SrEr2O4 [26], and SrYb2O4 [19] materials revealed that their ordering temperatures are all below 1 K, and for SrDy2O4 [20], no transitions to long-range order have been observed. All the compounds also show signs of short-range correlations in the bulk properties. This short-range order has been further investigated using neutron diffraction measurements, and these have revealed a rich variety of low-temperature, low-dimensional magnetic behavior [19,22,26–28]. The application of a magnetic field induces a variety of transitions in all of the SrL2O4 compounds [28]. The crystals are highly anisotropic, and for SrEr2O4, SrHo2O4, and SrDy2O4, plateaux in the magnetization curves at approximately one third of the saturation value appear for certain values of the applied field. Such features are usually indicative of the stabilization of a collinear two-spins-up one-spin-down (uud) structure.
In this paper, we report on the low-temperature properties of SrGd\(_2\)O\(_4\) single crystals, which have been grown for the first time using the floating zone technique and examined using susceptibility \(\chi(T)\), magnetization \(M(H)\), and specific heat \(C(T)\) and \(C(H)\) measurements. The magnetic Gd\(^{3+}\) ions have different electronic properties compared to the other lanthanides (as they are almost isotropic in the ground state where the orbital angular momentum is zero), and hence the magnetic behavior of SrGd\(_2\)O\(_4\) is expected to be markedly different to the other members of the Sr\(_2\)La\(_2\)O\(_4\) series. Our preliminary measurements of \(\chi(T)\) and \(M(H)\) on a powder sample of SrGd\(_2\)O\(_4\) indicated that there are transitions in this compound at 2.73 K in \(\chi(T)\) and around 19 kOe in \(M(H)\), contrary to what has been reported in Ref. [17].

These transitions were probably missed in the previous investigation as a result of having too large a step size in field, \(H\), and temperature, \(T\), during the data collection. Low-temperature measurements of \(\chi(T)\) on single-crystal samples for the fields applied along each of the principal axes show that below 2.73 K the most dramatic changes in the susceptibility occur when the field is applied along the \(c\) axis. Low-temperature \(M(H)\) data indicate that the magnetization processes along the \(a\) and \(b\) axes are quite uneventful, but that there are several in-field transitions when \(H\) is applied along the \(c\) axis. For \(H \parallel a\) and \(H \parallel b\), a single boundary is identified that delineates the transition from a low-field, low-temperature magnetically ordered regime to a high-field, high-temperature paramagnetic phase. Specific heat measurements on the single crystals of SrGd\(_2\)O\(_4\) were first performed in zero field, and these indicate that in addition to the transition seen at 2.73 K, another transition at an even lower temperature of 0.48 K is observed. \(C(T)\) in several applied fields and \(C(H)\) at a range of temperatures were also measured, and the results of all the bulk property measurements have been used to construct an \(H-T\) phase diagram of SrGd\(_2\)O\(_4\). Multiple magnetic phases have been identified, and this suggests that the magnetic ordering scheme in SrGd\(_2\)O\(_4\) is quite complex and rather different to that of the other Sr\(_2\)La\(_2\)O\(_4\) compounds. This is likely to be due to the fact that in the ground state the Gd\(^{3+}\) ions have zero orbital angular momentum. Hence crystal-field splittings are expected to be less important in SrGd\(_2\)O\(_4\), making it the best candidate in the Sr\(_2\)La\(_2\)O\(_4\) series for a realization of a classical Heisenberg exchange antiferromagnet. Dipole-dipole interactions are expected to be the leading perturbations in the Hamiltonian. SrGd\(_2\)O\(_4\) thus provides an interesting comparison and furthers the study of the influence of the spin-spin and spin-orbit interaction on the physics of the Sr\(_2\)La\(_2\)O\(_4\) [29] and similar systems, such as Ba\(_2\)La\(_2\)O\(_4\) [30–32].

### II. EXPERIMENTAL DETAILS

Single-crystal samples of SrGd\(_2\)O\(_4\) were grown using the floating zone method, similar to the procedure reported in Ref. [18]. Initially, powder SrGd\(_2\)O\(_4\) samples were prepared from high purity (99.99%) starting compounds SrCO\(_3\) and Gd\(_2\)O\(_3\), in an off-stoichiometric ratio of 1 : 0.875 (as it was found from x-ray diffraction measurements that when powders were mixed in a stoichiometric ratio the final materials contained a large, ∼15%, impurity phase of Gd\(_2\)O\(_3\)). Following the procedure described for the earlier syntheses of SrGd\(_2\)O\(_4\) [14,17], the powders were ground together and fired at ambient pressure in air at 1350°C for a total of 48 hours in an alumina crucible. In general, one intermediate grinding was used to ensure homogeneity of the mixtures. The purity of the SrGd\(_2\)O\(_4\) powder was verified by performing a Rietveld refinement [33] using x-ray diffraction data collected at room temperature, and allowing for two phases in the material—the desired SrGd\(_2\)O\(_4\) and an impurity phase of Gd\(_2\)O\(_3\). The results suggest that the SrGd\(_2\)O\(_4\) sample that was prepared starting with an off-stoichiometric SrCO\(_3\)/Gd\(_2\)O\(_3\) ratio is 99.80% pure SrGd\(_2\)O\(_4\), with \(\chi^2 = 1.158\) obtained for the fit. The atomic positions for SrGd\(_2\)O\(_4\) (within the orthorhombic space group \(Pnam\)) are given in Table I together with the refined unit cell parameters.

### TABLE I. Refined crystal structure parameters for SrGd\(_2\)O\(_4\) at room temperature.

<table>
<thead>
<tr>
<th>Atom</th>
<th>(x)</th>
<th>(y)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sr</td>
<td>0.7506(5)</td>
<td>0.6489(4)</td>
</tr>
<tr>
<td>Gd1</td>
<td>0.4270(4)</td>
<td>0.1127(3)</td>
</tr>
<tr>
<td>Gd2</td>
<td>0.4161(4)</td>
<td>0.6110(3)</td>
</tr>
<tr>
<td>O1</td>
<td>0.220(3)</td>
<td>0.181(2)</td>
</tr>
<tr>
<td>O2</td>
<td>0.135(3)</td>
<td>0.479(3)</td>
</tr>
<tr>
<td>O3</td>
<td>0.510(3)</td>
<td>0.785(2)</td>
</tr>
<tr>
<td>O4</td>
<td>0.423(4)</td>
<td>0.420(2)</td>
</tr>
</tbody>
</table>
The SrGd$_2$O$_4$ powder was subsequently isostatically pressed into rods of ~7 mm diameter and ~80 mm in length, and sintered in air at 1100 °C for 24 hours. Since no previous SrGd$_2$O$_4$ crystals existed, to start the growth, a polycrystalline rod was used as a “seed.” A high-temperature optical furnace (Crystal Systems Inc. Optical Floating Zone Furnace Model FZT-12000-X-VI-VP) equipped with four Xe arc lamps focused by four ellipsoidal mirrors was used. The feed and seed rods were counter-rotated at 10–20 rpm, and growth speeds ranging from 3 to 6 mm h$^{-1}$ were used. Several atmospheres were tried for the growth of SrGd$_2$O$_4$, but the best results were obtained for growths carried out in air at ambient pressure. The SrGd$_2$O$_4$ crystals grown in this manner were transparent to light, but were not wholly crack free, although large single-crystal regions could be isolated.

Once the crystal was produced, several samples were aligned using a backscattering x-ray Laue method and cut into thin rectangular prisms with faces perpendicular to the principal crystal axes. The samples varied from 10 to 15 mg for magnetization and magnetic susceptibility to 0.14 to 12.5 mg for specific heat measurements. Demagnetization corrections were applied following Aharoni [34].

A Quantum Design SQUID magnetometer was used to measure the magnetic susceptibility and magnetization along each of the principal crystallographic directions (with the applied field within an estimated 3° accuracy of the principal axes) in the ranges of 0.5 < $T$ < 400 K and 0 < $H$ < 70 kOe. The temperature range was extended below 1.8 K using an iQuantum [IQ2000-AHS-2RSO $^3$He system] refrigerator insert [35]. Additional higher-field (up to 100 kOe) magnetization measurements were performed using an Oxford Instruments vibrating sample magnetometer (VSM) with the lowest achievable temperature of 1.4 K.

Specific heat measurements were performed using a Quantum Design calorimeter, with the addition of a $^3$He insert, and were carried out both in zero field in the temperature range of 0.4 < $T$ < 400 K, and for the fields of up to 90 kOe applied along the $c$ axis. The heat capacity of single-crystal samples of SrY$_2$O$_4$ and SrLu$_2$O$_4$, which are nonmagnetic and isostructural to SrGd$_2$O$_4$, allowed for an estimation of the lattice contribution to the specific heat.

### III. RESULTS AND DISCUSSION

#### A. Temperature dependence of the magnetic susceptibility

**1. High-temperature limit**

The top panel of Fig. 2 shows the magnetic susceptibility versus temperature in 1 kOe, for a field applied along each of the three principal axes for a single-crystal sample of SrGd$_2$O$_4$. The bottom panel of Fig. 2 presents the temperature dependence of the inverse susceptibility. Here, the data are fitted to straight lines in the temperature range of 100 to 400 K. The parameters of these fits, the Weiss temperatures $\theta_{CW}$ and the calculated effective moments per magnetic ion $\mu_{eff}$, are listed in Table II. The average value of $\mu_{eff}$ is relatively close to the theoretical limit of 7.94 $\mu_B$, predicted for the free ion using Hund’s rules. The average value of $\theta_{CW}$ is consistent with $\theta_{CW} = -10.4(1)$ K, which is obtained from our measurements made on a powder sample of SrGd$_2$O$_4$, and this is comparable to the value of $-9.0(6)$ K obtained by Karunadasa, et al. [17].

There are no large differences in the Weiss temperatures or the effective moments for the three principal crystal directions of SrGd$_2$O$_4$, contrary to what is observed for the other Sr$_2$O$_4$ compounds [19,21]. It is likely that for all the Sr$_2$O$_4$ compounds other than SrGd$_2$O$_4$, the differences in the high-temperature susceptibility curves are due to the effects of low-lying crystal-field levels [24]. For SrGd$_2$O$_4$, however, in the ground-state gadolinium has a pure spin magnetic

<table>
<thead>
<tr>
<th>$H$</th>
<th>$\mu_{eff}$ ($\mu_B$)</th>
<th>$\theta_{CW}$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$H \parallel a$</td>
<td>8.03(8)</td>
<td>-11.0(6)</td>
</tr>
<tr>
<td>$H \parallel b$</td>
<td>8.10(5)</td>
<td>-7.4(2)</td>
</tr>
<tr>
<td>$H \parallel c$</td>
<td>7.88(2)</td>
<td>-12.3(1)</td>
</tr>
</tbody>
</table>

The value of the effective moment is close to 7.94 $\mu_B$ [36], which is calculated using Hund’s rules for a Gd$^{3+}$ ion.

FIG. 2. (Color online) (Top) Magnetic susceptibility vs temperature in an applied field of 1 kOe in the temperature range of 6 to 400 K for the field applied along the three principal axes of a single-crystal sample of SrGd$_2$O$_4$. (Bottom) Reciprocal of the molar susceptibility vs temperature and the least-squares regression fits to the data (using the Curie-Weiss model).
moment, and thus there is no distortion of the spherical $4f$ charge density due to the spin-orbit coupling, and hence no corresponding crystal-field anisotropy.

2. Low-temperature limit

The low-temperature susceptibility measurements on the single-crystal samples of SrGd$_2$O$_4$ are presented in Fig. 3 for a field of 100 Oe applied along the $a$, $b$, and $c$ directions. Here, a small cusp in the susceptibility at 2.73 K for the $a$ and $b$ axes, and a large decrease $\chi(T)$ when the field is applied along the $c$ axis suggests the presence of a phase transition, with the region of interest highlighted in the inset to Fig. 3. This is the first indication of a transition to a long-range ordered state identified for this compound, and the phase transition temperature appears to be much higher than that recorded for the other members of the Sr$L_2$O$_4$ series of compounds.

Around the transition temperature, in low applied fields such as 100 Oe, there is only a slight difference between the data obtained on warming after cooling in field (FC) and the zero-field-cooled warming (ZFC) regimes, which suggests that the magnetic susceptibility of SrGd$_2$O$_4$ is not particularly sensitive to sample history.

The lowest reachable experimental temperature when measuring magnetic susceptibility with the aid of a $^3$He probe is $\sim 0.5$ K. Unfortunately, this is not low enough to observe the second transition seen at 0.48 K in zero field for a powder sample of SrGd$_2$O$_4$ using heat capacity (see Sec. III C). However, the susceptibility data, acquired at low fields (such as 0.1 kOe in Fig. 3), along all of the principal axes of SrGd$_2$O$_4$ show an upturn in $\chi(T)$ at the lowest temperatures, hinting to the presence of a second transition in SrGd$_2$O$_4$ at temperatures just below 0.5 K.

The temperature dependencies of the magnetic susceptibilities in a range of fields applied along the $a$ and $b$ axes of SrGd$_2$O$_4$. There seems to be very little difference in the behavior at low fields and upon increasing the applied field along these axes. (Bottom) Low-temperature magnetic susceptibility obtained in the temperature range of 0.5 to 5 K, and field range of 0.10 to 50 kOe, with the fields applied along the $c$ axis of SrGd$_2$O$_4$. The transition seen at 2.73 K is suppressed by the application of higher fields, and in fields between 20 and 30 kOe a second feature in the magnetic susceptibility is observed. In fields above 30 kOe, again, only a single cusp in the low-temperature susceptibility is visible. This suggests a rich $H$-$T$ phase diagram for the fields applied along this axis of SrGd$_2$O$_4$, and this will be discussed in Sec. III D.
B. Field dependence of the magnetization

The field dependence of the magnetization and its derivative obtained with the field applied along the principal axes of SrGd$_2$O$_4$ at 0.5 K are shown in Fig. 5. In lower fields, the data collected for $H \parallel a$ shows no features in $dM/dH$, which remains small and practically flat. A similar behavior is observed for $H \parallel b$. In higher fields, both for $H \parallel a$ and $H \parallel b$, the magnetization deviates from a nearly straight line and shows signs of tending towards saturation, but being limited to a maximum field of 70 kOe (which after taking into account demagnetizing effects becomes even lower) it is difficult to accurately define the critical fields, therefore the error bars for the critical fields are relatively large. However, we have been able to clearly observe the magnetic saturation transitions for the field applied along either $a$ or $b$ axes of SrGd$_2$O$_4$ at temperatures above 1.4 K taking advantage of the higher fields available in the VSM (data not shown). Upon raising the temperature, the saturation transition moves gradually to lower fields and disappears completely above 2.73 K. The field dependence of the magnetization and its derivative $M(H)$ and $dM/dH$ data collected using the VSM for fields applied along either the $a$ or $b$ axes are combined with the SQUID data to construct the phase diagram discussed in Sec. III D.

The magnetization process of SrGd$_2$O$_4$ with $H \parallel c$ (the easy axis for this material) is more complicated, with three in-field transitions seen for data collected at 0.5 K. The initial rise in the magnetization is accompanied by a maximum in $dM/dH$ at $H_{c1} \approx 20.0$ kOe, and then for a small region of the applied field the magnetization shows much slower growth (with a minimum in $dM/dH$ seen at $\sim 22$ kOe), and then another small rise up to a second maximum in $dM/dH$ at $H_{c2} \approx 23.3$ kOe. Thus it is possible that the narrow region between $H_{c1}$ and $H_{c2}$, with an average magnetization value of $2.3 \mu_B$ (which is equal to roughly a third of the saturation magnetization value observed with $H \parallel c$), corresponds to a magnetization plateau. Such a plateau can be a sign of a field induced stabilization of a collinear two-spins-up-one-spin-down (uud) magnetic structure, in which on each triangle of spins, two are pointing up along the field and the third spin pointing down antiparallel to the field direction. Similar, albeit more distinct, plateaux have been observed in the magnetization curves of the other Sr$L_2$O$_4$ compounds [21], and thus, the stabilization of the uud spin structure could be a common feature of this family of materials. A third field induced transition for $H \parallel c$ is observed at $H_{c3} \approx 53.4$ kOe, and in even higher applied fields the magnetization shows signs of approaching saturation. The demagnetization corrections only have a small impact on the position of the field-induced phase transitions at $H_{c1}$ and $H_{c2}$ (of under 2%), but a somewhat larger effect on the position of $H_{c3}$ (of $\sim 5\%$). These field induced transitions have been confirmed across multiple independently aligned SrGd$_2$O$_4$ samples and their positions in field remain robust. Also, no hysteresis with the applied field along any of the principal crystal axes of SrGd$_2$O$_4$ is observed, as the magnetization data collected upon increasing and decreasing field coincide.

To further investigate the region of the applied field around $H_{c1}$ and $H_{c2}$ with $H \parallel c$ for SrGd$_2$O$_4$, $M(H)$ data were collected at several temperatures, and this and the derivative of the magnetization are shown in Fig. 6. The double peak in the derivative of magnetization seen in the data collected at 0.5 K becomes only a single peak at higher temperatures, and this single maximum in $dM/dH$ is initially shifted up, and then down in field as the temperature is increased. Since the two peaks in $dM/dH$ at $H_{c1}$ and $H_{c2}$ split from a single peak as the temperature is lowered, and with the extra data points obtained for plotting out the phase diagram for $H \parallel c$ (see Sec. III C), it seems plausible that at even lower temperatures the uud state would govern a larger region of the applied field.

The magnetization processes in all of the Sr$L_2$O$_4$ compounds are highly anisotropic. In SrEr$_2$O$_4$, the magnetic anisotropy is of pronounced easy-plane type [18,21], while in SrHo$_2$O$_4$ and SrDy$_2$O$_4$ an Ising-type behavior is observed (with different directions of an easy-axis on two different crystallographical sites for the magnetic ions) [21,24]. In SrYb$_2$O$_4$, the magnetization measured with $H \parallel c$ is much higher than in perpendicular directions, but the magnetic moments seem to have significant components along all three main symmetry axes [19]. In SrGd$_2$O$_4$, which is not affected by crystal-field splitting, almost all of the nontrivial magnetic behavior occurs for fields applied along the $c$ axis and it is possible that this direction is an effective easy axis of magnetization. The fact that the low-temperature

![FIG. 5. (Color online) (Top) Magnetization curves obtained with the field applied along the principal axes of SrGd$_2$O$_4$ at 0.5 K in the range of 0 to 70 kOe. (Bottom) Field derivatives of the magnetization. The field induced transitions are indicated by dashed lines at $H_{c1} = 20.0$ kOe, $H_{c2} = 23.3$ kOe, and $H_{c3} = 53.4$ kOe. Between $H_{c1}$ and $H_{c2}$, when $H \parallel c$, a narrow plateau in $dM/dH$ is observed at approximately one third of the value for the maximum moment along the $c$ axis.](094421-5)
magnetization below \( H_{c1} \) is significantly large (well above 1 \( \mu_B \) per \( \text{Gd}^{3+} \)) implies that the low-field magnetic structure in \( \text{SrGd}_2\text{O}_4 \) is noncollinear.

An interesting parallel to the behavior of \( \text{SrGd}_2\text{O}_4 \) may be found by looking at the \( \text{Gd} \) pyrochlore titanate. A Heisenberg-type antiferromagnet, \( \text{Gd}_2\text{Ti}_2\text{O}_7 \) orders magnetically at about 1 K \[37\]. Despite the absence of a significant single ion anisotropy for the \( \text{Gd}^{3+} \) ions, \( \text{Gd}_2\text{Ti}_2\text{O}_7 \) shows a highly anisotropic behavior in applied fields at lower temperatures \[38\], with multiple field-induced transitions \[39\]. In Sec. III D, we will further extend the analogy between the high-field behavior of \( \text{SrGd}_2\text{O}_4 \) and \( \text{Gd}_2\text{Ti}_2\text{O}_7 \), after first discussing the relevant \( C(H)/T \) results below.

### C. Specific heat

The temperature dependence of the specific heat, \( C(T) \), of single crystals of \( \text{SrGd}_2\text{O}_4 \) and of the nonmagnetic isostructural compounds \( \text{SrLu}_2\text{O}_4 \) and \( \text{SrY}_2\text{O}_3 \) were measured in zero field. The \( C(T)/T \) curves are shown in Fig. 7. Two \( \lambda \) anomalies, which correspond to transitions to long-range magnetic order, are observed at \( T_{N1} = 2.73 \text{ K} \) and \( T_{N2} = 0.48 \text{ K} \). The lattice contribution to the specific heat of \( \text{SrGd}_2\text{O}_4 \) can be estimated by measuring the heat capacity of two single-crystal nonmagnetic isostructural compounds: \( \text{SrY}_2\text{O}_3 \) and \( \text{SrLu}_2\text{O}_4 \). At temperatures below \( \sim 6 \text{ K} \), the lattice contribution to the specific heat of \( \text{SrGd}_2\text{O}_4 \) is negligible compared to the magnetic contribution, and thus the magnetic entropy can be estimated by integrating the \( C(T)/T \) curve, which has been extended linearly down to \( T = 0 \text{ K} \). It should be noted that, for a powder sample, measurements of the specific heat were also made using a dilution insert for the PPMS, which extended the temperature range of the zero field measurements down to 0.05 K (data not shown). These measurements confirmed that below \( T_{N2} \), \( C(T)/T \rightarrow 0 \) in a linear fashion as \( T \rightarrow 0 \text{ K} \), and that no more low-temperature transitions are present in the material down to 0.07 mK. The entropy curve for the single-crystal measurements is also shown in Fig. 7, and the (blue) right-hand axis should be used for the appropriate scale. It appears that by 6 K the full magnetic entropy expected for the \( \text{Gd}^{3+} \), \( J = 7/2 \), system is recovered.

From magnetic susceptibility and magnetization data, it has become apparent that for the fields applied along the \( a \) and \( b \) axes, \( \text{SrGd}_2\text{O}_4 \) shows no signs of any additional field-induced transitions apart from the transition from an ordered phase into a fully polarized state. With this in mind, the specific heat was measured for the fields applied along the \( c \) axis in a broad range of both fields and temperatures. The data for \( C(T)/T \) in several applied fields are shown in the top panel of Fig. 8. The two phase transitions in \( \text{SrGd}_2\text{O}_4 \), whose temperature is defined by the \( \lambda \) anomalies, seen in \( H = 0 \text{ Oe} \) move closer together upon increasing the strength of the applied field. The transitions “merge” into a single broad peak seen when \( C(T) \) is measured in 22.5 kOe, and after subsequent increases of the field this single peak is first shifted to higher and then lower temperatures. \( C(H)/T \) was also measured for \( H \parallel c \), and the data is shown in the bottom panel of Fig. 8. At the lowest measured temperature, two peaks are visible in the heat capacity data. As the temperature is increased, these peaks...
FIG. 8. (Color online) (Top) Temperature dependence of the specific heat divided by temperature of SrGd$_2$O$_4$ in several fields for $H \parallel c$. The two peaks associated with the transitions move closer with higher applied fields, before merging and then this single peak is shifted to lower temperatures in the highest applied fields. (Bottom) Field dependence of the specific heat divided by temperature at several temperatures for $H \parallel c$. At the lowest temperature, two peaks are observed in $C(H)$. As the temperature is raised, the peaks move closer together and eventually merge. For even higher temperatures, this single peak is suppressed and moves to lower fields.

two peaks move closer together, and in a small temperature range (between 1.2 and 1.3 K) are quickly suppressed in intensity before merging together at temperatures around 1.5 K. Subsequent increases in the temperature just shift this single peak to lower fields. All of the specific heat data collected for $H \parallel c$ are also used to construct the $H \parallel c$ phase diagram for SrGd$_2$O$_4$, which is described below.

The results of heat capacity measurements as a function of both temperature and magnetic field give direct information about the boundaries of the phase transitions observed in SrGd$_2$O$_4$, and these measurements demonstrate that SrGd$_2$O$_4$ is very different to other Sr$L_2$O$_4$ compounds. For SrGd$_2$O$_4$, magnetic susceptibility and specific heat measurements indicate that at least two transitions take place in zero-field at low temperatures, at 2.73 and 0.48 K, and no other Sr$L_2$O$_4$ compound measured so far shows two separate transitions to different long-range orders in zero applied field. Specific heat measurements, on SrDy$_2$O$_4$ [20], SrHo$_2$O$_4$ [40], SrEr$_2$O$_4$ [26], and SrYb$_2$O$_4$ [19] indicate the presence of short-range correlations developing at temperatures much higher than the observed transition temperatures in these materials. In the specific heat data collected for SrGd$_2$O$_4$, however, short-range correlations are limited to a small temperature range just above the transition temperature of 2.73 K. In contrast to SrGd$_2$O$_4$, the magnetic entropy contribution recovered at relatively low temperatures in SrEr$_2$O$_4$ [26], SrYb$_2$O$_4$ [19], and SrDy$_2$O$_4$ [20] amounts to $R \ln 2$, while in SrHo$_2$O$_4$ it is $R \ln 5$ [40], although this value is obtained over a much wider temperature range and without subtracting the nuclear Schottky anomaly.

D. $H$–$T$ phase diagram of SrGd$_2$O$_4$

By collating all of the susceptibility, magnetization, and specific heat data for $H \parallel c$, a magnetic field–temperature phase diagram may be constructed for SrGd$_2$O$_4$, and this is shown in the main panel of Fig. 9 [41]. The phase diagram reveals the rich and complex magnetic behavior of SrGd$_2$O$_4$ when $H \parallel c$. In addition to the paramagnetic regime, four other phases may be identified by looking at the boundaries on the diagram. This behavior is contrasted with a somewhat simpler response observed on application of magnetic field in directions orthogonal to $c$. For $H \parallel a$ and $H \parallel b$, the inset to Fig. 9 shows how the upper-temperature transition $T_{N1}$ gradually decreases in increasing fields. In this case, the magnetization measurements (limited to temperatures above 0.5 K) seem to indicate a single transition from a magnetically ordered to a disordered phase. It might be interesting to extend the magnetization measurement to the temperatures below $T_{N2} = 0.48$ K and check how the lowest-temperature...
phase responds to the application of field along all three crystallographical axes.

For all three directions of an applied field, the value of the upper-field transition $H_{c3}$ extrapolated to zero temperature returns approximately 60 kOe. Treated as a saturation field, this value could potentially be used to estimate the strength of the antiferromagnetic exchange interactions $J_{ij}$ provided that the number of interacting neighbors is known. In the absence of such information and because of the possible competition between different exchange links involved, neither $H_{sat}$ nor the $T_{CW}$ can be reliably converted to estimate $J_{ij}$, but what can be stated is that the energy of the exchange interactions in SrGd$_2$O$_4$ amounts to just a few degrees Kelvin. If this is the case, then it is rather obvious that given the relatively short Gd$^{3+}$–Gd$^{3+}$ separation (3.48 Å along the c axis) and a large magnetic moment ($J = 7/2$), dipolar interactions, which only for the nearest neighbors amount to roughly a Kelvin in units of temperature, cannot simply be disregarded—they must be taken into account as a leading perturbation to a purely exchange Hamiltonian.

In zero field, two transitions take place in SrGd$_2$O$_4$. The magnetic structures of the different phases remain unknown, but at least for the intermediate temperature regime, a nonzero susceptibility and magnetization rule out a simple collinear ordering. With the application of a field along the c axis of SrGd$_2$O$_4$, at the lowest temperature, three field-induced transitions are observed at $H_{c1}$, $H_{c2}$, and $H_{c3}$. It remains to be confirmed that the phase between $H_{c1}$ and $H_{c2}$ corresponds to the order observed in other SrLa$_2$O$_4$ compounds [21].

An interesting observation to make is on the nature of the high-field phase (above $H_{c3}$) at low temperatures usually labeled as a “fully-polarized state.” The magnetization continues to grow at a significant rate above $H_{c3}$ (see Fig. 5) and the magnetic heat capacity is also far from zero (the bottom panel in Fig. 8 shows that the heat capacity approaches zero only in much stronger fields around 80 kOe). This behavior is reminiscent of what has been observed in another Gd$^{3+}$ containing compound, the pyrochlore titanate. In particular, the field dependence of the heat capacity divided by temperature, $\theta_{CW}$, can be reliably converted to estimate $\theta_{CW}$, but what can be stated is that the energy of the exchange interactions are similar.

The data illustrate that there is a large difference between the magnetic behavior of SrGd$_2$O$_4$ and that of the SrHo$_2$O$_4$, SrEr$_2$O$_4$, and SrDy$_2$O$_4$ compounds investigated previously, even though the positions of the magnetic ions and the strength of the exchange interactions are similar. This is not totally unexpected as the low-temperature properties of SrGd$_2$O$_4$ have to be quite different compared to the other SrLa$_2$O$_4$ compounds in which the spin-orbit coupling and crystal-field anisotropies play a much more important role. An interesting point for further research is to establish the hierarchy of the magnetic interactions in SrGd$_2$O$_4$, in particular, the relationship between the exchange interactions and the dipolar forces and their influence on the selection of the ground state in this frustrated antiferromagnet.

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