Magnetic ordering in single-crystalline SrLaFeO$_4$ and Sr$_{1.1}$La$_{0.9}$FeO$_4$

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We report on the magnetic properties of single-crystalline SrLaFeO$_4$ and Sr$_{1.1}$La$_{0.9}$FeO$_4$. Both compounds crystallize in the tetragonal K$_2$NiF$_4$ structure ($I4/mmm$) with the La and Sr atoms randomly distributed over the 4e crystallographic sites of this structure. SrLaFeO$_4$ orders antiferromagnetically below 350 K, and Sr$_{1.1}$La$_{0.9}$FeO$_4$ orders antiferromagnetically at about 150 K. Both compounds exhibit magnetic structures described by orthorhombic $Cmca$-type symmetry and two magnetic propagation vectors $q_1=(1\over 2,1\over 2,0)$ and $q_2=(-1\over 2,1\over 2,0)$, representing the two possible magnetic domains. A collinear antiferromagnetic arrangement of the Fe$^{3+}$ moments confined to the tetragonal basal plane is found. Bulk magnetic studies on SrLaFeO$_4$ show that the susceptibility and Fe$^{57}$-Moessbauer studies. 5,6 Initially, the function of La concentration has been studied by magnetic transitions at lower temperatures (30 and 90 K). Low-temperature neutron diffraction data were taken to gain some insight into the nature of these transitions. © 2005 American Institute of Physics. [DOI: 10.1063/1.1861416]

Transition-metal oxides of the composition $A_2B$O$_4$ ($A$: rare earth and/or alkaline metal; $B$: transition metal) exhibit a wide variety of exotic properties that often occur in the proximity of the metal–insulator transition. 5 Among the fascinating properties, the concomitant evolution of the magnetism in some of these oxides remains one of the unsolved problems. Some attention has focused on the $3d$ transition-metal oxides based on Co$^{2+}$, Fe$^{3+}$ and Mn$^{4+}$, all of which are known to be charge-transfer insulators. In particular, the seemingly peculiar development of the magnetism in the Sr$_{2-x}$La$_x$FeO$_4$ system has received much recent interest. The magnetic properties of polycrystalline Sr$_{2-x}$La$_x$FeO$_4$ as a function of La concentration has been studied by magnetic-susceptibility and Fe$^{57}$-Moessbauer studies. 5,6 Initially, the substitution of La for the Sr in SrFeO$_4$ leads to a suppression of the antiferromagnetic ordering. However, the antiferromagnetism recovers for higher La concentrations at $x=0.7$ with a Néel temperature ($T_N$) of about 150 K. Up to $x=0.9$ (Sr$_{1.1}$La$_{0.9}$FeO$_4$), only a moderate increase of $T_N$ to about 160 K is found. For $x=1.0$ (SrLaFeO$_4$), however, $T_N$ is raised above room temperature.

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Already in 1980, Soubeyroux et al. 7 published the magnetic structure of SrLaFeO$_4$ determined by means of neutron powder diffraction. These authors established a collinear antiferromagnetic arrangement of Fe$^{3+}$ moments that lie in the tetragonal basal plane of SrLaFeO$_4$. Recently, various groups succeeded to successfully grow single crystals for some Sr$_{2-x}$La$_x$FeO$_4$ compositions. 8,9 For single crystalline SrLaFeO$_4$ the $c$-axis susceptibility displays additional anomalies at around 90 and 30 K. 9 Similarly, the $c$-axis magnetization reveals a small, but pronounced metamagnetic transition for fields around 1.8 T at 5 K. 10

Here, we focused on the magnetic properties of SrLaFeO$_4$ and Sr$_{1.1}$La$_{0.9}$FeO$_4$. In particular, we were interested to determine whether these two compounds exhibit the same moment configuration, even though their ordering temperature is vastly different. Furthermore, we wanted to shed some light on the nature of the additional anomalies around 30 and 90 K in SrLaFeO$_4$.

Single crystals of SrLaFeO$_4$ and Sr$_{1.1}$La$_{0.9}$FeO$_4$ were grown by the floating-zone method, details of the crystal growth are given in Ref. 9. Our single crystals of SrLaFeO$_4$ and Sr$_{1.1}$La$_{0.9}$FeO$_4$ had platelet shapes with approximate dimensions of $2 \times 2 \times 0.5$ and $4 \times 5 \times 1$ mm$^3$, respectively.
Our initial neutron diffraction studies used the single crystal diffractometer at the Los Alamos Neutron Science Center (LANSCE) at Los Alamos National Laboratory (Los Alamos NM). We collected 17 different histograms (covering the full reciprocal space) at room temperature for SrLaFeO₄, and six different histograms at 100 K, 200 K and at room temperature were collected for Sr₁.₁La₀.₉FeO₄.

Both compounds were found to crystallize in the tetragonal K₂NiF₄ structure (space group: I₄/mmm). It should be noted that we observed additional (half-indexed) peaks for SrLaFeO₄ that are not accounted for by the I₄/mmm crystal structure. These peaks are magnetic in origin. For Sr₁.₁La₀.₉FeO₄, on the other hand, the additional half-indexed reflections were observed only at 100 K. The Rietveld refinement program package GSAS I I was used to determine structural parameters SrLaFeO₄ and Sr₁.₁La₀.₉FeO₄ at 300 and 100 K, respectively. Exemplary contour plots for both crystals at respective temperatures are shown in Figs. 1 and 2. The magnetic peaks can be indexed with respect to the nuclear I₄/mmm structure.

As mentioned above, additional half-indexed magnetic reflections appear in the Laue pattern of SrLaFeO₄ and Sr₁.₁La₀.₉FeO₄ at 300 and 100 K, respectively. Exemplary contour plots for both crystals at respective temperatures are shown in Figs. 1 and 2. The magnetic peaks can be indexed with two different propagation vectors $q_1 = (\frac{1}{2}, \frac{1}{2}, 0)$ and $q_2 = (-\frac{1}{2}, \frac{1}{2}, 0)$ representing two possible domains. Assuming fully developed Fe³⁺ moments (4.6 $\mu_B$), we were able to achieve good fits of the magnetic intensities for both compositions when assuming two domains of orthorhombic Cmca symmetry. Thus, both compounds consists of antiferromagnetically coupled Fe³⁺ moments that are confined to the tetragonal basal plane (see Fig. 3). The here reported moment configuration is in excellent agreement with the powder-diffraction results on SrLaFeO₄.

Next the c axis and in-plane magnetization of the SrLaFeO₄ single crystal was studied using the 20-T superconducting magnet the Pulse Field Facility, National High Field Laboratory, at Los Alamos. The results of our magnetization studies at 2 K are shown in Fig. 4.

For magnetic fields applied within the tetragonal basal plane ($H \perp c$), there is a very slight S shape around 3–5 T (barely visible in Fig. 4), followed by a linear increase of the magnetization up to the highest field applied (18 T). Although not pronounced, we believe that the S shape is a signature of metamagnetic transition observed by Kawanaka et al. for fields applied along the (1,1,0) direction of one magnetic domain.

For magnetic fields applied along the c axis, on the other hand, the low-field susceptibility is much higher and a tendency toward saturation for fields above 12 T is observed. The low value for the saturation magnetization of about 0.16 $\mu$/Fe-atom indicates that strong in-plane antiferromagnetic coupling prohibits any substantial canting of the moments out of the tetragonal basal plane.

Finally, we used neutron diffraction to study the temperature dependence of the $(\frac{1}{2}, \frac{1}{2}, 1)$ magnetic reflection for SrLaFeO₄, in order to check for the possibility of additional

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TABLE I. Refined structural parameters for SrLaFeO₄ at room temperature. Space group: I₄/mmm. Lattice parameters: $a = 3.873(1)$ Å, $c = 12.738(1)$ Å.

FIG. 1. Exemplary contour plot of neutron diffraction data in reciprocal space for SrLaFeO₄ at room temperature. Half-indexed reflections are magnetic in origin. The peaks are indexed with respect to the nuclear I₄/mmm structure.

FIG. 2. Exemplary contour plot of neutron diffraction data in reciprocal space for Sr₁.₁La₀.₉FeO₄ at 100 K. Half-indexed reflections are magnetic in origin. The peaks are indexed with respect to the nuclear I₄/mmm structure.
magnetic transitions and their possible origin. We chose to study the \( \frac{1}{2}, \frac{1}{2}, 1 \) reflection because it will have contributions due to only one of the possible domains (the other domain has only parallel moment components). The experiments were performed on the ASTERIX spectrometer at LANSCE, Los Alamos National Laboratory, in a temperature range from 10 to 400 K. Figure 5 displays the temperature dependence of integrated magnetic \( \frac{1}{2}, \frac{1}{2}, 1 \) intensity normalized to one of the nuclear \( 1,1,2 \) peak, \( I_{\frac{1}{2}, \frac{1}{2}, 1}/I_{1,1,2} \). This way, contributions from thermal fluctuations due to the Debye–Waller factor are expected to cancel. A first observation is that the \( \frac{1}{2}, \frac{1}{2}, 1 \) disappears for temperatures above 350 K, indicating that the Néel temperature for our SrLaFeO\(_4\) single crystal is about 350 K, which agrees well with previous reports.\(^5\)\(^7\) Upon decreasing temperature, we first observe a smooth order-parameter-like increase of the \( \frac{1}{2}, \frac{1}{2}, 1 \) intensity, until there are two additional small jumps for the intensity at about 90 and 30 K. These temperatures coincide well with two characteristic temperatures observed in the magnetic susceptibility for \( H \perp c \).\(^9\) However, the increase in the integrated intensity is relatively small (\( \sim 6–8\% \) at either transition) at both temperatures. This observation immediately excludes a scenario, for which the sample transforms into a single-domain sample at low temperatures. Keeping in mind that the \( \frac{1}{2}, \frac{1}{2}, 1 \) tests only one of the two possible domains, one would expect that its intensity either doubles or drops to zero upon formation of a single-domain sample. Of course, some small domain redistribution cannot be excluded on the basis of our present data. On the other hand, some claims by Soubeyroux et al.\(^7\) suggest a more intriguing scenario. These authors speculated that SrLaFeO\(_4\) exhibits an “inhomogeneous distribution” of two collinear representations. They concluded that only about 92% of their sample displayed a moment configuration as shown in Fig. 4, while the other 8% exhibits a different collinear configuration of magnetic moments where the Fe moment at \( \frac{1}{2}, \frac{1}{2}, \frac{1}{2} \) points in the opposite direction. At present, we have little idea whether there might be any intrinsic mechanism causing such magnetic inhomogeneity, but it seems to explain the size of the jump for the integrated intensities at low temperatures.

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