Collinear antiferromagnetism in trigonal SrMn$_2$As$_2$ revealed by single-crystal neutron diffraction

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Abstract
Iron pnictides and related materials have been a topic of intense research for understanding the complex interplay between magnetism and superconductivity. Here we report on the magnetic structure of SrMn$_2$As$_2$ that crystallizes in a trigonal structure ($P\overline{3}m1$) and undergoes an antiferromagnetic (AFM) transition at $T_N = 118(2)$ K. The magnetic susceptibility remains nearly constant at temperatures $T \lesssim T_N$ with $H//c$ whereas it decreases significantly with $H//ab$. This shows that the ordered Mn moments lie in the $ab$ plane instead of aligning along the $c$-axis as in tetragonal BaMn$_2$As$_2$. Single-crystal neutron diffraction measurements on SrMn$_2$As$_2$ demonstrate that the Mn moments are ordered in a collinear Néel AFM phase with $180^\circ$ AFM alignment between a moment and all nearest neighbor moments in the basal plane and also perpendicular to it. Moreover, quasi-two-dimensional AFM order is manifested in SrMn$_2$As$_2$ as evident from the temperature dependence of the order parameter.

Keywords: neutron diffraction, magnetic structure, corrugated honeycomb structure, ordered magnetic moment, quasi-two-dimensional magnetic correlation

(Some figures may appear in colour only in the online journal)

1. Introduction

The recent discovery of unconventional superconductivity (SC) in Fe pnictides has led to an intense research effort aimed towards understanding their fundamental properties and the underlying mechanisms that lead to strong correlations between the lattice, charge and magnetic degrees of freedom [1–3]. Similar to the layered cuprate superconductors, the SC in FeAs-based compounds seems to arise close to an antiferromagnetic (AFM) phase, suggesting that magnetism and SC are closely intertwined in these systems [4–8]. However, the parent cuprates are insulators with strongly-correlated localized magnetic moments while the parent FeAs-based superconductors are metals with itinerant moments [1, 4, 9]. MnAs$_2$-based systems form a bridge between the high-$T_c$ cuprates and the FeAs-based materials, such as in tetragonal BaMn$_2$As$_2$ that orders with a G-type AFM structure and shares the same $I4/mmm$ crystal structure as many of the Fe pnictides but manifests an insulating ground state with localized moments similar to cuprates [10, 11]. Interestingly, unlike BaMn$_2$As$_2$, the insulator SrMn$_2$As$_2$ crystallizes in a trigonal unit cell (space group $P\overline{3}m1$) with a corrugated honeycomb structure [12–14] which yields a possibly frustrated Mn spin-system [15–17]. This kind of system attracts a lot of attention because depending upon the strength and nature of the spin interactions, different magnetic structures from a collinear Néel AFM phase to a helical and even to a stripe phase with alternating ferromagnetic (FM) stripes are possible [15, 16]. Moreover, SrMn$_2$As$_2$ lies in the realm of the Zintl compounds

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with CaAl$_2$Si$_2$-type structure like SrZn$_2$Sb$_2$ and YbZn$_2$Sb$_2$, which are extensively studied for high performance thermoelectrics [18, 19].

Magnetic susceptibility measurements on SrMn$_2$As$_2$ suggest that the spins order antiferromagnetically in the $ab$ plane, possibly with a helical AFM propagation vector [14]. Here we conclusively report from our single-crystal neutron diffraction studies that SrMn$_2$As$_2$ orders in a long-range collinear Néel AFM phase below $T_N = 118.2$ K with an ordered moment of $3.6(2)$ $\mu_B$/Mn ion at $T = 5$ K. Analysis of the magnetic order parameter further provides insight into the quasi two-dimensional character of the magnetic coupling. The two Mn moments within the unit cell lie in the $ab$ plane, forming a bilayer with antiparallel spins and can be viewed as a corrugated honeycomb lattice. High-resolution x-ray diffraction measurements were also performed to study the influence of magnetoelastic coupling in this system but no distortion of the lattice was observed down to a base temperature of 6 K within the resolution limit.

2. Experimental

Single crystals of SrMn$_2$As$_2$ were grown out of Sn flux using conventional high-temperature solution growth techniques [14, 20]. Both magnetic susceptibility $\chi(T)$ and neutron diffraction measurements were carried out on the same single crystal of mass 44 mg, with dimensions $4 \times 3 \times 1$ mm$^3$. The crystals grow as flat plates with the $c$-axis perpendicular to them. A Quantum Design, Inc., superconducting quantum interference device magnetic properties measurement system was used for the $\chi(T)$ measurements. Energy-dispersive x-ray measurements on the same single crystal confirmed the composition to be a pure 122 phase. Single-crystal neutron diffraction measurements were performed at the thermal triple-axis spectrometer, TRIAX, at the University of Missouri Research Reactor. Measurements were carried out with an incident energy of 14.7 meV, using Soller collimations of $60^\circ$–$40^\circ$-sample-$40^\circ$–$80^\circ$. Pyrolytic graphite filters were placed both before and after the sample to reduce higher-order wavelengths. The sample was mounted on the cold finger of a closed-cycle helium cryostat to reach temperatures of 5 K $\leq T \leq 300$ K. The lattice parameters were measured to be $a = b = 4.29(1)$ Å and $c = 7.24(1)$ Å at 5 K. Rocking scans performed through the Bragg peaks showed a full width at half maximum of $\approx 0.3^\circ$ confirming the good mosaicity of the sample. High-energy x-ray diffraction measurements were performed on a 1 mg single crystal at the 6-ID-D station at the Advanced Photon Source using an x-ray wavelength of $\lambda = 0.123712$ Å and a beam size of $100 \times 100$ mm$^2$. The sample was cooled down using a closed-cycle He cryostat. Two Be domes were placed over the sample and evacuated, and a small amount of He exchange gas was subsequently added to the inner dome for thermal equilibrium. An aluminized-Kapton heat shield also surrounded the sample and inner Be dome. The cryostat was mounted on a 6-circle diffractometer and a MAR345 image plate was used to measure the diffracted x-rays [21].

Figure 1. Temperature dependence of magnetic susceptibility $\chi(T)$ for $H \parallel c$ and $H \perp c$ with $H = 1000$ Oe. Two views of the magnetic structure are shown as insets. Right inset: a side view of the collinear AFM alignment of the Mn atoms at two different heights along the $c$-axis marked by blue (Mn1) and red (Mn2) planes respectively, forming a corrugated honeycomb layer, with two antiparallel spin directions. Left inset: the ordered Mn moments in each unit cell as viewed along the $c$-axis.

3. Results

The $T$-dependence of the magnetic susceptibilities $\chi(T)$ with applied magnetic field ($H$) along the $c$-axis ($\chi_c$, $H \parallel c$) and perpendicular to it ($\chi_{ab}$, $H \perp c$) are shown in figure 1. For $H \perp c$, $\chi_{ab}$ starts to decrease rapidly below $T_N = 118.2$ K with a distinct change in the slope while $\chi_c$ remains almost constant suggesting an antiferromagnetic (AFM) transition with the ordered moments aligned within the $ab$ plane. The right inset of figure 1 shows the crystal structure of SrMn$_2$As$_2$ in the hexagonal setting, consisting of corrugated honeycomb [Mn$_2$As$_2$]$^-$ layers stacked along the $c$-axis separated by Sr$^{2+}$ layers.

Theoretical calculations predict that frustration in a corrugated honeycomb lattice can result in a helical or an incommensurate magnetic ground state [15, 16]. Therefore, neutron diffraction measurements were performed to determine the magnetic structure. As a first step, we searched for possible magnetic propagation vectors by surveying the reciprocal space for magnetic Bragg peaks. Experiments were carried out in two configurations—first in the ($h 0 l$) horizontal scattering plane followed by measurements in the ($h h l$) scattering plane. Extensive measurements along the three principal directions [001], [$h 0 0$], and [$h h 0$] found no additional magnetic scattering beyond the magnetic contributions at the Bragg peaks corresponding to the chemical crystal structure as shown in figure 2. Additional diffraction peaks observed in figure 2(c) correspond to the scattering from the aluminum (Al) sample holder which was confirmed by rocking scans through the Al peak positions with no significant observed variation in peak intensities.

As shown in figure 3, magnetic intensity develops only at the nuclear Bragg peak positions below $T_N$ suggesting
that the magnetic unit cell is same as that of the chemical unit cell with \( q = 0 \) AFM structure. This is built by the two magnetic Mn atoms within the same unit cell as marked by red and blue spheres/planes respectively, in the insets to figure 1. Symmetry analysis using the program SARAh-Representational Analysis [22] provided distinct magnetic structures for this system which includes collinear FM or AFM alignments with the moments aligned either along the \( c \)-axis or in the \( ab \)-plane. No canting of the ordered moments is allowed by symmetry, thus excluding any possible canting of the ordered moment out of the \( ab \)-plane.

Since we observe magnetic intensity in \( l00 \) (reflected in \( l = 0 \)) type Bragg peaks in our neutron diffraction measurements, we can confirm that the spins are not aligned along the \( c \)-axis. This is due to the fact that the neutron scattering cross-section is sensitive only to the component of the ordered magnetic moment perpendicular to the scattering vector. Moreover, no indication of a FM signal was found in magnetization measurements [14]. The only remaining possibility is a collinear Néel AFM phase with the \( 180^\circ \) AFM alignments between a moment and all nearest neighbor moments as shown in the insets to figure 1.

All accessible peaks in the \((h 0 l)\) and \((h h l)\) scattering planes have been measured and analyzed with a total of 14 independent peaks, excluding peaks with high non-magnetic intensities. Figures 3(a)–(c) show the temperature dependence of the integrated intensities of the \((002), (100)\) and \((112)\) peaks. Rocking scans at specific temperatures are shown in figures 3(d)–(f). There are no changes in the peak intensities between 120K and 140K but as the temperature is lowered below \( T_N = 118 (2) K \), the magnetic contribution sets in and the peak intensities increase.

The magnetic moment can be calculated from the integrated intensities measured by rocking scans on a series of peaks in both the \((h 0 l)\) and \((h h l)\) configurations. The magnetic intensity is given by [23]

\[
I_M = N_M \frac{(2\pi)^3}{\nu_M} \sum_{G_M} \delta(Q - G_M) |F_M(G_M)|^2,
\]

where \( \nu_M \) is the magnetic unit cell volume, \( N_M \) is the number of such cells in the sample, \( Q \) is the scattering vector and \( G_M \) is the magnetic reciprocal-lattice vector. \( F_M \) is the magnetic structure factor and is given by

\[
F_M(G_M) = \frac{\gamma_0}{2} \sum_{j} g_j f_j(S_j \cdot \delta Q) [G_M \cdot \delta Q - W^{-1}],
\]

where \( \gamma = 1.193 \) is the magnetic dipole moment of the neutron in units of nuclear Bohr magneton, \( r_0 = 2.818 \times 10^{-15} m \) and \( g \) is the spectroscopic splitting factor. \( f(G_M) \) and \( e^{-W} \) are the magnetic form factor and the Debye–Waller factor for the magnetic Mn ions, respectively. The index \( j \) is a sum over all \( j \)th Mn ions in the unit cell. The quantity \( S_j \) is the component of \( S \) perpendicular to \( Q \) which contributes to the magnetic scattering amplitudes. The magnetic intensities were obtained from the difference in integrated intensities between 5K and 140K (\( T_N \)) data sets. The difference is normalized with the corresponding integrated intensities at 140K which are purely of nuclear origin. The temperature dependence of the nuclear intensity is almost constant above \( T_N \) as evident from figures 3(a)–(c). Thus any excess contribution of nuclear intensity at 5K due to the Debye–Waller factor can be neglected.

Though the ordered collinear Mn moments lie in the \( ab \) plane of the trigonal lattice, it is not possible to uniquely...
and was determined to be 3.6(2) at 0 K, which is close to the experimental value of 0.58(1) determined from $\chi_{ab}$ and $\chi_c$ at $T = 2$ K in figure 1. The minor difference is probably due to a deviation from our assumption of equal domain population, since the in-plane anisotropy is expected to be small due to the $s$-state nature of the Mn$^{2+}$. Ion with a 3d electronic configuration. An estimate of an exemplary domain population with domain ratios of 0.24 : 0.38 : 0.38 or 0.28 : 0.28 : 0.44 for the three high symmetric directions can give a value of 0.58 for $\chi_{ab}/\chi_c$, which was used in estimating the error in the ordered moment from unequal domain population and was found to be less than 5% and within the quoted error bar.

In the following, we analyze the detailed temperature dependence of the magnetic contribution ($\chi_M$) to the Bragg peaks which is proportional to the square of the ordered moment. For $T$ close to $T_N$, it is predicted to have a power law behavior with a critical exponent $2\beta$ given by $\chi_M = \chi_0 (1 - T/T_N)^{2\beta}$ [30]. Figures 3(a)–(c) show the power-law fit of the measured magnetic intensity for $T > 90$ K. The critical exponent $\beta$ was determined for six independent peaks as shown for the three peaks in figures 3(a)–(c) and was consistently found to be 0.21(3) which lies between the values expected for a three-dimensional (3D) Heisenberg spin system ($\beta_{3D,Heisenberg} = 0.36$) and that for a purely 2D Ising/XY system which predict critical exponents of $\beta_{2D,Ising} = 0.125$ and $\beta_{2D,XY} = 0.13$, respectively [30]. Thus SrMn$_2$P$_2$ appears to behave like a quasi-2D AFM system which is also consistent with magnetic susceptibility studies above $T_N$ [14].

This quasi-2D behavior can be explained qualitatively from the magnetic and lattice structures. Within the unit cell, the two antiparallel Mn spins, Mn1 and Mn2, lie on different planes perpendicular to $c$, forming a corrugated honeycomb layer (figure 1 insets) with a nearest-neighbor distance of 3.06(1) Å. The distance between the nearest Mn atoms in the layers in the adjacent unit cells along the $c$-axis is the much larger distance of 6.05(1) Å. This difference is expected to result in much weaker interplanar exchange interactions compared to intraplanar interactions, leading to a quasi-2D

**Figure 3.** Integrated intensities of the (002), (100) and (112) Bragg peaks as a function of temperature $T$ are shown in (a)–(c) respectively. The solid line is a power law fit given by, $I_M = \chi_0 (1 - T/T_N)^{2\beta}$ for $T > 90$ K. (d)–(f) Rocking scans at specific temperatures (as mentioned in panel (e)) around (002), (100) and (112) peaks, respectively. The data at 120 K and 140 K overlap with each other.

determine their orientation in this plane due to the symmetry of the crystal structure and possible domain orientations. Considering equal domain populations and the magnetic moments aligned along the high symmetry directions, there are six possible spin directions (or domains) for the Mn moment: $S = \pm [1 0 0]$, $\pm [0 1 0]$ and $\pm [1 1 0]$. The second Mn moment within the magnetic unit cell is aligned in the corresponding antiparallel direction. If $\eta_i$ is the angle between $S_i$ and $Q$ for the $i$th domain, then the average magnetic interaction vector contributing to the magnetic intensity for that particular peak is given by $\langle |S_i|^2 \rangle = S^2(1 - \langle \cos^2 \eta_i \rangle)$. The ordered moment is obtained by $\mu_B \chi_M$ and was determined to be 3.6(2) $\mu_B$/Mn at 5 K. The quoted error includes the uncertainty in the domain populations explained later. The ordered moment suggests local moment AFM behavior as observed in BaMn$_2$As$_2$ [10]. It is somewhat lower than the nominal 5.0 $\mu_B$/Mn expected for the high-spin state of Mn$^{2+}$ but is comparable to other Mn-122 compounds like BaMn$_2$As$_2$ [10], BaMn$_2$P$_2$ [24], SrMn$_2$P$_2$ [24], CaMn$_2$Bi$_2$ [25], and CaMn$_2$Sb$_2$ [17, 26, 27]. The reduced moment can be attributed to the strong spin-dependent hybridization between the Mn 3d and the As 4p orbitals as shown by density functional calculations [28] and to the expected quasi-two-dimensionality of the Mn–Mn spin interactions (see below). This is, however, different from the Fe-122 compounds where the Fe moment is greatly reduced due to the itinerant nature of the magnetism [1, 3, 9].

Based on the proposed equal domain configuration, we now calculate the expected ratio between $\chi_{ab}$ and $\chi_c$, assuming negligible magnetic anisotropy. If $\theta$ is the angle between the spin direction and the applied field, the susceptibility is given by $\chi_{\theta} = \chi_0 \cos^2 \theta + \chi_{\perp} \sin^2 \theta$, where $\chi_{\parallel}$ and $\chi_{\perp}$ are the susceptibility parallel and perpendicular to the applied field [29]. For $H||e$, $\theta = 90^\circ$ for all the domains and thus $\chi_c = \chi_{\perp}$. To simplify the calculation for $\chi_{ab}$, we further assume $H||[1 0 0]$ for the in-plane susceptibility measurement. Then equal domain populations with $\theta = 0^\circ$, 60$^\circ$ and 120$^\circ$ result in $\chi_{ab} = \chi_{\parallel}/2$ at $T = 0$ ($\chi_{\parallel} = 0$ at $T = 0$ for a collinear AFM). Therefore, $\chi_{ab}/\chi_c = 0.5$ at $T = 0$ which is close to the experimental value of 0.58(1) determined from $\chi_{ab}$ and $\chi_c$ at $T = 2$ K in figure 1. The minor difference is probably due to a deviation from our assumption of equal domain population, since the in-plane anisotropy is expected to be small due to the $s$-state nature of the Mn$^{2+}$ ion with a 3d electronic configuration. An estimate of an exemplary domain population with domain ratios of 0.24 : 0.38 : 0.38 or 0.28 : 0.28 : 0.44 for the three high symmetric directions can give a value of 0.58 for $\chi_{ab}/\chi_c$, which was used in estimating the error in the ordered moment from unequal domain population and was found to be less than 5% and within the quoted error bar.
spin system. It has been shown by Mermin and Wagner [31] that a 2D arrangement of Heisenberg spins cannot order long-range except at $T = 0$. Therefore, interlayer coupling and/or anisotropic effects are necessary to induce long-range AFM order. If these are weak, one expects a value of $\beta$ to be less than the 3D Heisenberg value of 0.36. Experimentally, one observes $0.1 < \beta \leq 0.25$ for a range of quasi-2D spin systems [32], a regime that encompasses our value of $\beta = 0.21(3)$.

Additionally, we have performed high-energy x-ray diffraction measurements to study the strength of the magnetoelastic coupling in SrMn$_2$As$_2$. Magnetically-induced lattice distortions have been observed in several pnictide systems as in CaFe$_2$As$_2$ [33], and Ba(Fe$_1-x$Co$_x$)$_2$As$_2$ [34], where the lattice changes from the tetragonal ThCr$_2$Si$_2$-type structure to the $Fmmm$ orthorhombic phase as the magnetic moment direction breaks the tetragonal symmetry. Similarly, in SrMn$_2$As$_2$, collinear AFM Mn moments in the $ab$ plane break the trigonal symmetry and thus a lattice distortion is expected perpendicular and parallel to the moment directions. This would result in a change of lattice symmetry to a monoclinic C2/m phase with observable splitting of Bragg peaks in the $(h k 0)$ plane below $T_N$. However, no lattice distortion was observed in SrMn$_2$As$_2$ down to 6 K and an upper limit on any possible relative lattice distortion was determined to be $3 \times 10^{-4}$, as calculated from the experimental resolution. This can also be associated with its CaAl$_2$Si$_2$-type structure where the greater flexibility of the transition-metal $d$ orbitals allows for better adjustments of the lattice structure compared to the ThCr$_2$Si$_2$-type structure and thus a greatly reduced lattice distortion [35].

4. Conclusion

In conclusion, we have shown that SrMn$_2$As$_2$ orders in a collinear Néel AFM phase with the Mn spins aligned along the basal plane of the trigonal unit cell having a net ordered moment of $3.6(2) \mu_B$/Mn which is smaller than the full high-spin value of $5.0 \mu_B$/Mn. The magnetic interaction is quasi-two-dimensional with a strong in-plane exchange interaction within the corrugated honeycomb Mn layer compared to weak interplanar interaction between the layers. Lastly, the magnetoelastic coupling does not cause a measurable lattice distortion in the ordered state.

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