I. INTRODUCTION

Materials composed of two-dimensional (2D) layers, either in bulk compounds or reduced to a few or single layers, provide avenues for enhanced quantum phenomena. In this context 2D monolayer graphene, formed from the exfoliation of weakly connected van der Waals (vdW) bonded layers in graphite, has ignited widespread interest due to a variety of novel behavior. General 2D material research has extended beyond graphene to the broader class of cleavable vdW materials. A particular focus has been on materials with magnetic ions on 2D layers with predictions of topological and enhanced quantum behavior in the bulk compounds down to the monolayer. In the 2D atomic layer limit for Heisenberg spins the Mermin-Wagner theorem prohibits magnetic order, however, Onsager’s theorem for Ising spins supports the stabilization of long range 2D order, which would apply to materials with strong magnetocrystalline anisotropy. Therefore, an understanding of the exchange interactions and anisotropy provides indications on the stability and behavior of low dimensional magnetism.

The class of 2D vdW materials has begun expanding and includes $\text{Fe}_6\text{GeTe}_6$, $\text{CrXTe}_3$ ($X=$Si, Ge, Sn), $\text{CrX}_2\text{Te}_6$ ($X=$Si, Ge, Sn), $\text{CrPS}_4$, $\text{CrCl}_3$, and $\text{VTe}_2$. Ferromagnetism in atomically thin 2D layers has been shown in $\text{CrI}_3$ and $\text{CrGeTe}_3$. One of the initial classes of 2D vdW materials is $\text{MnPSe}_3$. $\text{MnPSe}_3$ has magnetic ions forming the same hexagonal motif as graphene with crystal space group R3 (\#148). Antiferromagnetic order of the Mn$^{2+}$ ions (3d$^5$), in the high-spin state $S=5/2$, has been reported at $74 \pm 2$ K from neutron scattering. The size of the magnetic and crystallographic unit cell are the same due to the propagation vector of $k=(000)$. The reduced $T_N$ compared to a Curie-Weiss temperature of -201 K supports the low dimensional nature of the magnetism. Considering the reduction to monolayer theoretically the magnetic ground state of 2D $\text{MnPSe}_3$ remains antiferromagnetic with a predicted ordering temperature of 88 K, close to the value of the bulk system. Experimental indications show that the magnetic ordering temperature remains similar from bulk to a few nanometer. Considering the magnetic structure in Ref. 36 for bulk $\text{MnPSe}_3$ concludes from neutron diffraction that the spins are confined to the $ab$-plane but cannot rule out a non-zero component along the $c$-axis. A more recent study using powder diffraction also concluded the spins for $\text{MnPSe}_3$ are confined to the basal plane. The moment direction has implications for an understanding of the anisotropic, Ising, XY or 3D Heisenberg nature of the spins, and any competition between single ion anisotropy and dipolar anisotropy.

Comparisons of $\text{MnPSe}_3$ and related $\text{MnPS}_3$ have led to intriguing findings which form broader context for this study. Despite the analogous Mn$^{2+}$ 2D lattice and similar magnetic ordering temperatures, $T_N(\text{MnPSe}_3)=74$ K and $T_N(\text{MnPS}_3)=78$ K, the magnetism is altered from predominantly isotropic in $\text{MnPS}_3$ to indications of unusually large XY anisotropy in $\text{MnPSe}_3$ along with a flipping of the spin direction. The magnetic anisotropy in $\text{MnPSe}_3$ was proposed as a consequence of the zero-field splitting of the Mn-ion ground state (S=5/2, L=0) from a combination of spin-orbit coupling and the axial crystal field due to a trigonal distortion of the MnS$_6$ octahedra. This octahedral structural distortion, however, is reduced in $\text{MnPSe}_3$ compared to $\text{MnPS}_3$. Indeed, there is debate in the literature regarding the origin of the anisotropy in $\text{MnPS}_3$. A spin-gap at the magnetic Bril-
louin zone center of 0.5 meV was measured using Inelastic Neutron Scattering (INS)\textsuperscript{42}. This was modeled within a framework of linear spin-wave theory with the introduction of an easy-axis term to account for the anisotropy, rather than a dipolar coupling term. It was noted that the calculated difference between the two routes was negligible for the data. One signature of the dipole-dipole coupling is a splitting of the magnon excitation\textsuperscript{43}. Indications of this was observed with the high resolution available for the data. One signature of the dipole-dipole interaction is best described as hosting both single-ion anisotropy and dipolar coupling, with continued research aimed at further insights, for example through Raman studies\textsuperscript{45}.

Here we present powder neutron diffraction and INS measurements on MnPSe$_3$ to access the anisotropy and exchange interactions in this bulk vdW layered material. Magnetic excitations, consistent with spin waves, are observed in the magnetically ordered regime that dampen but persist above T$_N$. Analysis of the data provides a model spin Hamiltonian that requires the inclusion of both intralayer interactions, $J_{iab}$, $J_{iab}$, $J_{iab}$ and interlayer exchange interaction $J_c$. The data does not reveal any observable spin-gap, indicating weak anisotropy. The results reveal that while the exchange interactions in MnPSe$_3$ are dominated by intralayer interactions, the appreciable interlayer ($J_c$) interactions contrast to that observed for MnPS$_3$, with an alteration of an order of magnitude.

II. EXPERIMENTAL DETAILS

A. Sample synthesis

Powdered samples of MnPSe$_3$ and MnPS$_3$ were synthesized through annealing of their constituent elements: Mn (99.95\%, Alfa Aesar), P (99.999\%+, Alfa Aesar), and Se (99.999\%, Alfa Aesar) for MnPSe$_3$, or S (99.9999\%, Puratronic) for MnPS$_3$. For each sample, under argon atmosphere to avoid oxygen and moisture effects, stoichiometric amounts of the elements were measured out to equal 6 g, and ground in an agate mortar and pestle until reaching a fine texture and uniform color. The fine powder was then loaded into a pellet press die and transferred to ambient atmosphere for pressing. Once compressed, the resulting pellet was sealed in a fused-quartz tube under a constant vacuum of 10-2 Torr, before being placed in a muffle furnace for annealing. To avoid over-pressure of the tube due to volatilization of P, S, and Se, the samples were slowly heated to 730\°C over 3 days, then held at that temperature for 1 week, before allowing to cool to room temperature. The resulting fine powders of bright green MnPS$_3$ and wine red MnPSe$_3$ were verified for correct crystal structure on an Empyrean X-ray diffractometer (Malvern Panalytical).

B. Neutron powder diffraction

Neutron powder diffraction was carried out on the HB-2A powder diffractometer at the High Flux Isotope Reactor (HFIR), Oak Ridge National Laboratory (ORNL)\textsuperscript{46,47}. A vertically focusing germanium monochromator was used to select a wavelength of 1.12 Å from the Ge(117) reflection and 1.54 Å from the Ge(115) reflection. HB-2A has a Debye–Scherrer geometry and the diffraction pattern was collected by scanning a 120° bank of 44° He detectors in 0.05° steps to give 2θ coverage from 5 to 130°. Soller collimators of 12° are located before each detector. Measurements with a 1.12 Å wavelength were collected at 295 K, 100 K, 75 K and 15 K for 4 hours. Measurements with a 1.54 Å wavelength were collected from 15 to 110 K in 5 K temperature steps for 1 hour. A 5 gram sample of MnPSe$_3$ was contained in a vanadium sample holder with diameter 0.9 cm. The data was normalized using a vanadium measurement. The wavelengths and detector positions were calibrated with a Si measurement. The neutron data collected was reduced using Mantid\textsuperscript{48}.

C. Inelastic neutron scattering

INS measurements were performed on the time-of-flight direct geometry spectrometer Sequoia\textsuperscript{49} at the Spallation Neutron Source, ORNL. Data were collected on MnPSe$_3$ and MnPS$_3$ powder samples. The samples were loaded into a 1 cm diameter cylindrical aluminium can and measured utilizing the 3-sample changer in the closed cycle refrigerator (CCR). An identical empty aluminium can was measured and used to subtract the background from the sample environment. Data were collected with incident energies of $E_i$=20 meV and $E_i$=8 meV. Both of these energy were in high resolution mode. For $E_i$=20 meV the Fermi chopper frequency was 240 Hz with a T$_0$ chopper frequency of 60 Hz giving an energy resolution at the elastic line of 0.48 meV. For $E_i$=8 meV the Fermi chopper frequency was 120 Hz with a T$_0$ chopper frequency of 30 Hz giving an energy resolution at the elastic line of 0.18 meV. The energy resolution was calculated using rez.mcvine.ornl.gov and confirmed by taking cuts of the elastic line in the data. The momentum resolution, Q, was based on fitting the resolution of the Bragg peaks at the elastic line. Measurements were collected for 6 hours at 200, 100, 70 and 15 K. Shorter measurements of 1 hour from 22 K to 95 K in steps of 7° were also collected with $E_i$=8 meV. The neutron data collected were reduced using Mantid\textsuperscript{48}. Data reduction and analysis utilized the DAVE software\textsuperscript{50} and modeling was performed with SpinW\textsuperscript{51}. 
III. RESULTS AND DISCUSSION

A. Neutron powder diffraction measurements

Neutron powder diffraction measurements were performed on MnPSe$_3$ to probe the crystal and magnetic structure as a function of temperature, as shown in Fig. 1. To initially investigate the crystal symmetry a short wavelength of 1.12 Å was used to give a large Q coverage up to 10 Å$^{-1}$. This allowed detailed measurements of the lattice, atomic positions and thermal parameters with Rietveld refinement, which was performed using the Fullprof software.$^{52}$ The refinements did not require the inclusion of any preferred orientation parameter. No indication of any structural symmetry lowering from the R3 ($\#148$) was found with these measurements, or with the measurements using the longer wavelength of 1.54 Å. MnPSe$_3$ consists of Mn-layers stacked in an ABC stacking sequence separated by 6.65 Å at 15 K. The Se layers are stacked in an ABAB sequence. The trigonally distorted MnSe$_6$ octahera have Mn-Se bond distances of 2.757(8)Å and 2.721(7)Å.

Turning now to the magnetic structure. As previously reported measurements indicated, the spins are in the ab-plane with a propagation vector $k=(000)^{36,40}$. We consider this further based on symmetry arguments using two complimentary approaches: Representational analysis and magnetic space groups. The representational analysis approach was performed using the SARAh software.$^{54}$ This assumes the high temperature paramagnetic space group remains unaltered in the magnetic phase. This is consistent with the refinements of the crystal structure at all temperatures measured, see Fig. 1(a). Under this case the irreducible representations (IRs) contain magnetic structures with spins either confined to the ab-plane or magnetic structures with spins along the c-axis, see Table I. To allow for a magnetic structure with a component of the spin out of either the basal plane or c-axis requires the combination of more than one IR. This goes against Landau theory which states that for a continuous transition only one IR is required. The IR $\Gamma_4$ was used to determine the magnetic structure in this approach. Conversely, using the magnetic space group approach we find no maximally allowed magnetic space group compatible with the data. The only maximally allowed magnetic space groups found with the Bilbao Crystallography Server$^{55}$ have spins only along the c-axis. Therefore we were required to go to a further lower symmetry subgroup to find the magnetic space group of P-1’ ($\#2.6$), in BNS notation. Within this magnetic space group the spins are symmetry-allowed to have non-zero and different magnetic components along the a-, b- and c-axis. Results from both the IR and magnetic space group approaches were refined against the data. No distinction was observed and therefore we follow the highest symmetry approach since no breaking of the a=b symmetry was measured. The magnetic structure therefore confines the spins to the ab-plane, with arbitrary direction within the plane. The refined magnetic moment was determined to be 3.83(3) $\mu_B$/Mn$^{2+}$. We note that the P-1’ magnetic space group allows both a definite spin direction within the ab-plane, as well as a component along the c-axis, moreover this space group supports magnetoelectric coupling. Further measurements will be of interest to probe for symmetry breaking within the layer to provide more insight into the magnetic structure.

FIG. 1. Refinements of neutron powder diffraction measurements on MnPSe$_3$ with a wavelength of $\lambda=1.12$ Å at (a) 15 K and (b) 295 K. (c) Refined Mn$^{2+}$ magnetic moment at 15 K to 110 K in 5K temperature steps through the magnetic ordering transition. The solid line is a fit to a power law. (d) Crystal and magnetic structure of MnPSe$_3$ produced using VESTA.$^{53}$ The unit cell is indicated by the solid lines.

| IR BV Atom | BV components |
|------------|---------------|
| $m_{||a}$ | $m_{||b}$ | $m_{||c}$ |
| $\Gamma_1$ $\psi_1$ | 1 | 0 | 0 | 1 |
| | 2 | 0 | 0 | 1 |
| $\Gamma_2$ $\psi_2$ | 1 | 0 | 0 | 1 |
| | 2 | 0 | 0 | -1 |
| $\Gamma_3$ $\psi_3$ | 1 | 1 | 0 | 0 |
| | 2 | 1 | 1 | 0 |
| $\Gamma_4$ $\psi_4$ | 1 | 1 | 0 | 0 |
| | 2 | -1 | -1 | 0 |
| $\Gamma_5$ $\psi_5$ | 1 | 1 | -1 | 0 |
| | 2 | 1 | -1 | 0 |
| $\Gamma_6$ $\psi_6$ | 1 | 1 | -1 | 0 |
| | 2 | -1 | 1 | 0 |

TABLE I. Basis vectors for the space group R3:H with $k=(000)$ in Kovalev’s notation. The decomposition of the magnetic representation for the Mn site (0, 0, .17484) is $\Gamma_{Mag}=1\Gamma_1^0+3\Gamma_1^1+3\Gamma_2^0+3\Gamma_2^1+6\Gamma_3^0+6\Gamma_3^1$. The atoms of the nonprimitive basis are defined according to 1: (0, 0, 0.16817), 2: (0, 0, 0.83183).
To follow the evolution of the magnetic ordering, the magnetic and crystallographic scattering were refined from measurements in 5 K steps from 15 to 110 K, shown in Fig. 1(c). No anomalies of the crystal lattice were observed within this temperature range. The temperature dependence of the ordered Mn$^{2+}$ moment was fit to a power law, shown in Fig. 1. The onset of long range order is consistent with the reported transition temperature of 74 K. The best fit critical exponent was $2\beta = 0.14(1)$. This is consistent with 2D ordering, as has been observed in related bulk vdW compounds.$^{36}$

**IV. INELASTIC NEUTRON SCATTERING MEASUREMENTS**

To access the magnetic exchange interactions and anisotropy, inelastic neutron scattering measurements were performed on MnPSe$_3$ at various temperatures through the $T_N = 74$ K magnetic ordering transition, see Fig. 2. Complimentary data, shown in Fig. 3, at the same incident energy and temperature conditions were collected for MnPS$_3$, $T_N = 78$ K, to allow for a direct comparison of the data between MnPSe$_3$ and MnPS$_3$. 

FIG. 2. INS data for MnPSe$_3$. $E_i=20$ meV Bose factor corrected data at (a) 15 K, (b) 40 K, (c) 70 K, (d) 100 K and (e) 200 K. $E_i=8$ meV Bose factor corrected data at (f) 15 K, (g) 40 K, (h) 70 K, (i) 100 K. (j) Bose corrected intensity in region $E=0.3-0.8$ meV and $Q=1.135-1.235$ Å$^{-1}$ to follow the evolution of the low energy scattering around the magnetic Bragg point.

FIG. 3. INS data for MnPS$_3$. $E_i=20$ meV at (a) 15 K, (b) 40 K, (c) 70 K, (d) 100 K and (e) 200 K. $E_i=8$ meV at (f) 15 K, (g) 40 K, (h) 70 K, (i) 100 K. (j) Bose corrected intensity in region $E=0.3-0.8$ meV and $Q=1.17-1.27$ Å$^{-1}$ to follow the evolution of the low energy scattering around the magnetic Bragg point.
The MnPS$_3$ scattering within the magnetically ordered phase, as expected, is consistent with the single crystal data in the literature$^{42}$. The data presented here allows the temperature dependence to be observed.

The data shown in Fig. 2 for the inelastic scattering in MnPSe$_3$ have temperature and Q dependence behavior consistent with magnetic scattering. The well defined excitations within the magnetically ordered regime broaden but remain robust at 100 K and even persist to 200 K, the highest temperatures measured. This temperature behavior indicates low dimensional exchange interactions and the presence of short range correlations to high temperatures. Contrasting the INS data for MnPSe$_3$ with MnPS$_3$ shows a reduced energy scale with a maximum energy of 8.5 meV for MnPSe$_3$ compared to 11.5 meV for MnPS$_3$, despite the similar ordering temperatures. Nevertheless, similar temperature behavior is observed for MnPS$_3$ and MnPSe$_3$, with well defined magnetic excitations dampening, but remaining present above the magnetic transition.

Focusing on low energy and inspecting the data at the magnetic Bragg positions (Q=1.2-1.3 Å$^{-1}$) reveals another difference in the spectra of MnPSe$_3$ as MnPS$_3$. The MnPS$_3$ inelastic scattering signal extends down to E=0.5 meV, see Fig. 3(a) and Fig. 3(f) and Ref. 42. For MnPSe$_3$ the low energy magnetic scattering is qualitatively different, with strong scattering cutting off below 2 meV. This can be seen most clearly in the S(Q, ω) scattering in Fig. 4(a) and constant momentum Fig. 4(b) and constant energy Fig. 4(c) cuts taken at low energy in MnPSe$_3$. While it is tempting to assign this behavior to a spin-gap due to increased anisotropy, as predicted from magnetic susceptibility measurements$^{41}$, closer inspection reveals weak scattering dispersing from the magnetic Bragg points at all energies. Therefore any spin-gap is smaller than the energy resolution. Instead, as we show below when the data are modeled, the magnetic excitation spectra indicates an alteration of the interlayer interactions between MnPSe$_3$ and MnPS$_3$.

In Fig. 2(h) and Fig. 3(h) the Bose corrected intensity in a fixed Q and energy transfer window are plotted to follow the low energy temperature dependence through T$_N$. The T<T$_N$ results are fit to $\chi''(T) \propto (-\Delta/k_B T)$. A pronounced change in the spectral weight occurs at the expected ordering temperatures consistent with T$_N$(MnPSe$_3$)=74 K and T$_N$(MnPS$_3$)=78 K.

A. Magnetic Exchange Interactions

To gain a quantitative understanding of the magnetic excitations we turn to modeling the data with linear spin wave theory to extract exchange interactions and anisotropy. Inelastic neutron scattering on single crystals is the premier method to achieve this due to the well understood scattering cross section S(Q, ω) and access to four dimensional energy and momentum space with Q=H,K,L. Indeed this was done on MnPS$_3$ and so
FIG. 6. (a) Exchange interactions between the Mn ions (purple spheres) in MnPSe$_3$ used in the spin Hamiltonian. Inelastic neutron scattering data and model based on the best fit exchange interaction parameters for (b) $E_i= 20$ meV and (c) $E_i= 8$ meV. All data was collected at 15 K. The boxes highlight the data cuts shown in the rest of the figure. (d) Cut along energy with $E_i=20$ meV in the range $1.12 \leq Q (\text{Å}^{-1}) \leq 1.22$. (e) Cut along energy with $E_i=8$ meV in the range $0.5 \leq E (\text{meV}) \leq 0.8$. (f) Cut along $Q$ with $E_i=8$ meV in the range $2.0 \leq E (\text{meV}) \leq 2.3$. (g) Cut along $Q$ with $E_i=8$ meV in the range $3.7 \leq E (\text{meV}) \leq 4.0$.

The fitting is not repeated here for MnPSe$_3$. The reproduced results are shown in Fig. 5 to confirm the validity of the model for the MnPS$_3$ powder sample. However, we note that for low dimensional vdW compounds the hierarchy of intralayer and interlayer exchange interactions can allow the extraction of detailed Hamiltonians from the $S(|Q|, \omega)$ powder data, as has been shown in previous studies$^{19,57-59}$. We therefore follow this approach to provide a spin Hamiltonian for MnPSe$_3$. Moreover, the high symmetry of MnPSe$_3$, as confirmed above with neutron powder diffraction, simplifies the required exchange parameter further and makes the development of a model spin Hamiltonian for MnPSe$_3$ more straightforward. Namely, MnPS$_3$ has a distorted hexagonal 2D layer which leads to further exchange interactions as compared to MnPSe$_3$, which has identical bond lengths in the layer.

The inelastic measurements presented revealed no spin-gap (Fig. 4) and consequently we do not consider any anisotropy in the Hamiltonian. This is consistent with the expected reduced single-ion anisotropy from the less distorted octahedral environment of the Mn ion. Moreover, the magnetic structure of spins confined to the ab-plane indicates reduced anisotropy. Our initial model was based on a 2D Hamiltonian on a honeycomb lattice,

$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j = \frac{1}{2} \sum_{i \neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$

that included $J_{1ab}$, $J_{2ab}$ and $J_{3ab}$ intralayer exchange interactions shown in Fig. 6(a). The $\frac{1}{2}$ prefactor has been included in the exchange interactions to be consistent with reported parameters in the literature$^{60-62}$ as well as the full spin ($S=5/2$). This 2D model, however, did not reproduce the observed INS scattering in MnPSe$_3$ at low energy. Therefore we extended the Hamiltonian to include interlayer exchange interactions ($J_c$) to give a 3D model isotropic Hamiltonian.

We begin with a confirmation that the previous reported results for MnPS$_3$ in Ref. 42 can be reproduced by the powder data. These measurements make the approximation of a higher symmetry in which the layers are undistorted with $J_1=0.77$ meV, $J_2=0.07$ meV, $J_3=0.18$ meV and $J_c=-0.0038$ meV. An easy-axis single ion anisotropy term of 0.0086 meV is also included to model the observed spin-gap in MnPS$_3$. Figure 5 shows the powder data are well reproduced using these parameters.

We now turn to modeling the exchange interactions in MnPSe$_3$, that have not been experimentally determined with INS. Exchange interactions based on a $J_{1ab}$-$J_{2ab}$-$J_{3ab}$ 2D model without an interlayer $J_c$ or
anisotropy terms have been theoretically reported in several studies\textsuperscript{60–63}. These considered the required antiferromagnetic structure of MnPSe\textsubscript{3} on the 2D hexagonal lattice with the hierarchy of $J_1 > J_2 > J_3$ and all interactions positive (antiferromagnetic). The different studies show broad agreement. Therefore after initial confirmation of these parameters, the focus of this study will be on the interlayer interactions, $J_c$.

Starting with the values reported in Refs. 60 and 61 gives very close agreement to the measured excitation energy range, within 1.5\%. We therefore applied a scale factor and kept these values fixed in the subsequent analysis at $J_{1ab}=0.45$ meV, $J_{2ab}=0.03$ meV and $J_{3ab}=0.19$ meV. To model the data, we compared measured and calculated $S(|Q|, \omega)$, as well as constant energy and momentum cuts. Simulations were performed with SpinW\textsuperscript{51} using 100000 random orientations per Q value. To rule out any single-ion anisotropy term being able to model the data, we applied this to a modified Hamiltonian; how-ever, the results did not show agreement. Therefore we extended the data, we applied this to a modified Hamiltonian; however, the results did not show agreement. Therefore we applied the $J_c$ term and performed least squares fitting to find the best-fit $J_c$ parameter to the data. The results are shown in Fig. 6. The measured INS data are best modeled with exchange interactions $J_{1ab}=0.45$ meV, $J_{2ab}=0.03$ meV, $J_{3ab}=0.19$ meV and $J_c=0.031(5)$ meV.

Comparing the exchange interactions we have determined for MnPSe\textsubscript{3} with those measured for MnPS\textsubscript{3} reveals an enhanced interlayer interaction energy in MnPSe\textsubscript{3} of an order of magnitude greater than reported for MnPS\textsubscript{3} from INS\textsuperscript{42}. This is coupled to a reduced energy scale for intralayer $J_{1ab} - J_{2ab} - J_{3ab}$ interactions in MnPSe\textsubscript{3} compared to MnPS\textsubscript{3}. While any anisotropy present in MnPSe\textsubscript{3} is beyond the limits of our measurement, adding single ion anisotropy to the spin Hamiltonian indicates that the energy is likely on the same scale or smaller than that measured in MnPS\textsubscript{3}. This suggests that interlayer coupling plays a more significant role than anisotropy in MnPSe\textsubscript{3} compared to MnPS\textsubscript{3}.

The extraction of a model spin Hamiltonian that best fits the INS data reveals MnPSe\textsubscript{3} as having dominant intralayer interactions with non-negligible interlayer interactions. Conversely, MnPS\textsubscript{3} is closer to the 2D limit in the bulk compound. This balance of competing interactions can explain the similar magnetic ordering temperatures, with lower intralayer exchange interactions in MnPSe\textsubscript{3} that drive a lower ordering temperature being offset by higher interlayer exchange interactions that stabilize long range three dimensional order in the bulk. Further theoretical and experimental investigations of MnPSe\textsubscript{3} from the bulk down to the monolayer, where $J_c = 0$, will be of interest to follow the crossover from the weakly three dimensional exchange interaction model to purely 2D in-plane interactions.

V. CONCLUSIONS

Neutron powder diffraction and inelastic neutron scattering measurements on MnPSe\textsubscript{3} allowed model magnetic exchange interactions to be extracted. A lack of observable symmetry lowering of the crystal structure or spin-gap in the inelastic excitation spectra is consistent with magnetic order confined to the $ab$-plane. The measured magnetic excitations show well-defined spin waves that were modeled within a linear spin wave theory. While the intralayer $J_{1ab} - J_{2ab} - J_{3ab}$ interactions dominate the magnetic behavior, extension beyond a 2D Hamiltonian was required with the inclusion of a non-negligible interlayer interaction $J_c = 0.031(5)$ meV to model the data.

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