Pressure induced magneto-structural phase transitions in layered RMn2X2 compounds (invited)

Shane Kennedy
ANSTO

Jianli Wang
University of Wollongong, jianli@uow.edu.au

Stewart Campbell
University of New South Wales, stewart.campbell@adfa.edu.au

Michael Hofmann
Technische Universitat Munchen

S X. Dou
University of Wollongong, shi@uow.edu.au

Follow this and additional works at: https://ro.uow.edu.au/aiimpapers

Part of the Engineering Commons, and the Physical Sciences and Mathematics Commons
Pressure induced magneto-structural phase transitions in layered RMn2X2 compounds (invited)

Abstract
We have studied a range of pseudo-ternaries derived from the parent compound PrMn2 Ge 2, substituting for each constituent element with a smaller one to contract the lattice. This enables us to observe the magneto-elastic transitions that occur as the Mn-Mn nearest neighbour distance is reduced and to assess the role of Pr on the magnetism. Here, we report on the PrMn2 Ge 2−xSix, Pr1−xYxMn2 Ge 2, and PrMn2−xFex Ge 2 systems. The pressure produced by chemical substitution in these pseudo-ternaries is inherently non-uniform, with local pressure variations dependent on the local atomic distribution. We find that concentrated chemical substitution on the R or X site (e.g., in Pr 0.5 Y 0.5 Mn 2 Ge 2 and PrMn 2 Ge 0.8 Si 1.2) can produce a separation into two distinct magnetic phases, canted ferromagnetic and canted antiferromagnetic, with a commensurate phase gap in the crystalline lattice. This phase gap is a consequence of the combination of phase separation and spontaneous magnetostriction, which is positive on transition to the canted ferromagnetic phase and negative on transition to the canted antiferromagnetic phase. Our results show that co-existence of canted ferromagnetic and antiferromagnetic phases depends on chemical pressure from the rare earth and metalloid sites, on local lattice strain distributions and on applied magnetic field. We demonstrate that the effects of chemical pressure bear close resemblance to those of mechanical pressure on the parent compound.

Keywords
compounds, rmn2x2, layered, transitions, invited, phase, pressure, structural, magneto, induced

Disciplines
Engineering | Physical Sciences and Mathematics

Publication Details
Pressure induced magneto-structural phase transitions in layered RMn 2 X 2 compounds (invited)
Shane Kennedy, Jianli Wang, Stewart Campbell, Michael Hofmann, and Shixue Dou

Citation: Journal of Applied Physics 115, 172617 (2014); doi: 10.1063/1.4870582
View online: http://dx.doi.org/10.1063/1.4870582
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/115/17?ver=pdfcov
Published by the AIP Publishing

Articles you may be interested in
Magnetoelastic coupling induced magnetic anisotropy in Co2(Fe/Mn)Si thin films

The tunable magnetostructural transition in MnNiSi-FeNiGe system

Magnetostructural phase transitions and magnetocaloric effects in MnNiGe1xAlx

Re-entrant ferromagnet Pr Mn 2 Ge 0.8 Si 1.2 : Magnetocaloric effect

Magnetic phase transitions in (Tb,Y)Mn 2 M 2 ( M = Ge and Si) systems
J. Appl. Phys. 93, 8185 (2003); 10.1063/1.1541652

Re-register for Table of Content Alerts
Create a profile. Sign up today!
Pressure induced magneto-structural phase transitions in layered $RMn_2X_2$ compounds (invited)

Shane Kennedy,$^{1, a)}$ Jianli Wang,$^{1, 2}$ Stewart Campbell,$^3$ Michael Hofmann,$^4$ and Shixue Dou$^2$

$^1$The Bragg Institute, Australian Nuclear Science and Technology Organisation, Lucas Heights, NSW, Australia
$^2$Institute for Superconductivity and Electronic Materials, University of Wollongong, Wollongong, NSW, Australia
$^3$School of Physical, Environmental and Mathematical Sciences, University of New South Wales, Canberra, ACT, Australia
$^4$Forschungs-Neutronenquelle Heinz Maier-Leibnitz, Technische Universität München, Garching, Germany

(Presented 7 November 2013; received 23 September 2013; accepted 29 October 2013; published online 15 April 2014)

We have studied a range of pseudo-ternaries derived from the parent compound PrMn$_2$Ge$_2$, substituting for each constituent element with a smaller one to contract the lattice. This enables us to observe the magneto-elastic transitions that occur as the Mn-Mn nearest neighbour distance is reduced and to assess the role of Pr on the magnetism. Here, we report on the PrMn$_2$Ge$_2$-$x$Si$_x$, Pr$_{1-x}$Y$_x$Mn$_2$Ge$_2$, and PrMn$_{2-x}$Fe$_x$Ge$_2$ systems. The pressure produced by chemical substitution in these pseudo-ternaries is inherently non-uniform, with local pressure variations dependent on the local atomic distribution. We find that concentrated chemical substitution on the $R$ or X site (e.g., in $Pr_{0.5}Y_{0.5}Mn$_2$Ge$_2$ and $PrMn$_2$Ge$_{0.8}Si$_{1.2}$) can produce a separation into two distinct magnetic phases, canted ferromagnetic and canted antiferromagnetic, with a commensurate phase gap in the crystalline lattice. This phase gap is a consequence of the combination of phase separation and spontaneous magnetostriction, which is positive on transition to the canted ferromagnetic phase and negative on transition to the canted antiferromagnetic phase. Our results show that co-existence of canted ferromagnetic and antiferromagnetic phases depends on chemical pressure from the rare earth and metalloid sites, on local lattice strain distributions and on applied magnetic field. We demonstrate that the effects of chemical pressure bear close resemblance to those of mechanical pressure on the parent compound. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4870582]

I. INTRODUCTION

The RMn$_2$X$_2$ series (where $R$ is a rare earth and $X$ is Si or Ge) are layered intermetallic compounds in which the $R$ and Mn atoms lie in alternate layers, separated by layers of $X$ atoms. These compounds display a fascinating array of magnetic phases, due to the strong dependence of the Mn-Mn nearest neighbour distance on magnetic and atomic structure, we learn a great deal about the nature of these compounds and about their potential for technological applications.

Here, we report our findings on the pseudo-ternaries, PrMn$_2$Ge$_{2-x}$Si$_x$ (Ref. 2), Pr$_{1-x}$Y$_x$Mn$_2$Ge$_2$ (Ref. 3), and PrMn$_{2-x}$Fe$_x$Ge$_2$ (Ref. 4), derived from the parent compound PrMn$_2$Ge$_2$, (in which the Mn-Mn nearest neighbour distance is 2.915 Å at room temperature), substituting for each constituent element with a smaller one to contract the lattice so as to observe the magneto-elastic transitions that occur as the Mn-Mn nearest neighbour distance is reduced and to assess the role of Pr on the magnetism.

II. EXPERIMENTAL DETAILS

The synthesis of the compounds discussed here, PrMn$_2$Ge$_{2-x}$Si$_x$, Pr$_{1-x}$Y$_x$Mn$_2$Ge$_2$, and PrMn$_{2-x}$Fe$_x$Ge$_2$, have been described in Refs. 4–6.

It suffices to note here that all samples studied had been subjected to long annealing at high temperatures (typically above 900 °C for at least 7 days) and were of high phase purity (98% or above).

Magnetization measurement, differential scanning calorimetry (DSC) measurements were performed at the Institute of Superconducting and Electronic Materials, University of Wollongong, in order to identify the magnetic phases and phase transition temperatures in these compounds.
Neutron diffraction measurements variously evaluated here have been obtained at ANSTO on Wombat, Echidna, and Medium Resolution Powder Diffractometer (MRPD), at HZ-Berlin on E6, at ISIS on GEM, and at FRM-II on Spodi. Synchrotron X-ray diffraction experiments were performed at the powder diffraction beamline (PD) at the Australian Synchrotron.

III. RESULTS AND DISCUSSION

A. Magnetic phases of PrMn$_2$Ge$_2$

The magnetic properties of PrMn$_2$Ge$_2$ have been well reported$^{7-9}$ and will not be covered in detail here. Sufficient to note that four magnetic phases have been observed, as depicted in Fig. 1, with transition temperatures of 105 K, 286 K, 330 K, and 420 K for Fmi + FPr, Fmi, Fmc, and AFl phases, respectively.

Primarily the magnetism is located in the planes of Mn atoms, which display planar antiferromagnetic order at all temperatures below $T_N$ (420 K) and an additional axial ferromagnetic component below the transition between AFl and Fmc phases ($T \sim 330$ K). Pr orders ferromagnetically (with moments parallel to the ferromagnetic component of the Mn moments) at $T_{CPD} \sim 105$ K.

Identification of these magnetic structures is made with neutron powder diffraction, as illustrated in Fig. 2, where we see a selected region of the diffraction pattern from PrMn$_2$Ge$_2$ between 4 K and 450 K. For convenience, the magnetic phase transitions are marked on the plot. Most of the transitions are clear from changes in the diffraction pattern, and we note, in particular, the change in the position of the magnetic satellites of the incommensurate Fmi phase, around the (101) and (103) reflections, signifying a decrease in the period of rotation of the magnetic moments about the c-axis with increasing temperature.

B. Pseudo-ternaries of PrMn$_2$Ge$_2$

Mixing of any of the three constituent atoms of PrMn$_2$Ge$_2$ with a smaller atom invariably reduces the interatomic spacings, leading to modification of the magnetic structure. Typically, at a critical interatomic spacing ($d_{Mn-Mn} = 2.86 \text{Å}$) where the axial component of the Mn-Mn magnetic exchange interaction changes sign, we observe a transition from Fmc to one in which the axial component of the Mn moment couples antiferromagnetically between planes (denoted by AFlmc). This change is independent of the nature of the substitution, being an essential feature of the electronic interactions between Mn atoms. We illustrate this point with the magnetic phase diagrams for Pr$_{1-x}$Y$_x$Mn$_2$Ge$_2$ (Fig. 3), and PrMn$_2$Fe$_x$Ge$_2$ (Fig. 4), where the magnetic phase boundaries are drawn as a function of a lattice parameter to highlight the importance of interatomic spacing between nearest neighbour Mn atoms. Here, the lines of thermal expansion for specific values of substituent are kinked near the boundary between AFlmc and Fmc and in the region where Pr orders ferromagnetically.

The electronic origin of the boundary between Fmc and AFlmc magnetic states has been established by first principles calculations of the electronic structure of the two magnetic states$^{2}$ using a spin polarized localized density approximation. Due to the anisotropic nature of the response of the lattice to changes in temperature and pressure, corresponding changes in the magnetic exchange energy of the two magnetic states (Fmc and AFlmc) have different dependencies. Specifically, the calculations indicate that in Pr$_{0.3}$Y$_{0.3}$Mn$_2$Ge$_2$, the Fmc state is favoured at ambient pressure below $\sim 50$ K and above $\sim 150$ K, whereas the AFlmc state is favoured between those temperatures, in agreement with our experimentally determined phase diagram (Fig. 3).

Experimentally, we observe that the boundary between Fmc and AFlmc is associated with a region of co-existence of both states.$^2$ In neutron diffraction studies of the PrMn$_2$Ge$_2$-$x$Si$_x$ series, we find that the transition from AFl to either Fmc or AFlmc is also accompanied by spontaneous magnetostriction, which is positive on transition to Fmc, and negative on transition to the AFlmc phase.$^2$ Because of this magneto-elastic coupling co-existence of these two magnetic states is accompanied by a structural separation, such that a large “gap” ($\Delta a/a \approx 0.5\%$) appears in the phase diagram of PrMn$_2$Ge$_2$-$x$Si$_x$. This gap, as shown in Fig. 3 of Ref. 2, appears to be temperature independent, extending all the way from 2 K to the transition to the AFl state.

Identification of a phase gap in PrMn$_2$Ge$_2$-$x$Si$_x$ prompts us to suggest that there will also be one in the shaded regions of Figs. 3 and 4 as well. Confirmation of this will require more detailed structural studies.

The appearance of a region of magnetic phase co-existence raises the question, as to why some crystallites

![Schematic of the magnetic structures found in PrMn$_2$Ge$_2$. Pr atoms are green, Mn atoms are maroon, and Ge atoms are grey.](image)
adopt the Fmc state whereas others adopt the AFmc state. It is pertinent to add here that, to our knowledge, co-existence is only found in pseudo-ternary compounds and ternary compounds where the rare earth ion is suspected to be in a mixed valence state (as shown in Fig. 5 of Ref. 2).

We know from diffraction studies that there is no hint of long range chemical ordering on the mixed lattice site. Whether the chemical distribution is random or non-random is unclear from neutron diffraction. Certainly, considerable local lattice strain fields could be expected. Short range chemical order may also be present, and even the possibility of a miscibility gap is not excluded. Indeed strain can be seen in the neutron diffraction from PrMn$_2$Ge$_2$$_x$Si$_{1-x}$, even though instrumental factors dominate the measured peak widths. Concentration dependent lattice and magneto-elastic strain components can be identified in the (110) and (002) reflections. The lattice component, which is largely independent of temperature, is seen in both (110) and (002) reflections while the magneto-elastic component is evident only in the (110) reflection at Ge/Si concentrations where Fmc and AFmc co-exist.

C. Magneto-structural anomalies

High resolution scattering studies are needed to accurately determine strain distributions, domain sizes and the potential role of atomic short range order.

Our high resolution X-ray powder diffraction study of the PrMn$_2$Ge$_2$$_x$Si$_{1-x}$ series of compounds reveals many details of the structural features of these pseudo-ternaries. Most notably, we clearly see that chemical order is present in the Ge/Si planes of all the pseudo-ternaries of this series, and that the lattice strain broadening is highly anisotropic. We illustrate this point in Fig. 5, which shows the (220) and (008) Bragg reflections from a pseudo-ternary (PrMn$_2$Ge$_{0.8}$Si$_{1.2}$) in which the two phase region is clearly seen. As in the neutron diffraction pattern of Fig. 2, the magnetic phase transitions are marked on the plot. Several features of note are: spontaneous magnetostriction can be seen at both the AF$_1$ $\leftrightarrow$ PM and Fmc/AFmc $\leftrightarrow$ AF$_1$ phase boundaries, the magnetostriction at

![FIG. 2. Selected region of neutron powder diffraction patterns from PrMn$_2$Ge$_2$, showing four magnetic phase transitions between 4 K and 450 K. (Data collected on the Wombat diffractometer at ANSTO.) The magnetic phases Fmi + FPr, Fmi, Fmc, AFl, and PM are depicted in Fig. 1.](image)

![FIG. 3. Magnetic phase diagram of Pr$_{1-x}$Y$_x$Mn$_2$Ge$_2$ as a function of a lattice parameter. Values of x are indicated above the line for Neel temperature (T$_N$). Thermal expansion for selected concentrations of Y is indicated by fine black lines. The dashed line shows the extrapolated boundary between Fmc and Fmi.](image)

![FIG. 4. Magnetic phase diagram of PrMn$_{2-x}$Fe$_x$Ge$_2$ as a function of a lattice parameter. Values of x are indicated above the line for Neel temperature (T$_N$). Thermal expansion for selected concentrations of Fe is indicated by fine black lines.](image)

![FIG. 5. Thermal dependence of the (220) (left) and (008) (right) X-ray Bragg peaks from PrMn$_2$Ge$_{0.8}$Si$_{1.2}$, showing spontaneous magnetostriction at the magnetic phase transitions, and phase separation in the Fmc/AFmc region. Magnetic labels as above in Fig. 1, except AFmc = AF$_1$ plus axial antiferromagnetic order. (Data collected on the PD beamline at the Australian Synchrotron.)](image)
the $AFmc \leftrightarrow AFl$ is of opposite sign to that of the $Fmc \leftrightarrow AFl$ phase boundary, the (220) reflection is substantially broader than the (008) reflection, and whereas the (220) reflection is completely resolved into two peaks in the mixed $Fmc/AFmc$ phase, the (008) reflection barely broadens. Thus, we see that one signature of the $AFmc$ phase is an increase in $c/a$ ratio. The variation in intensity of $Fmc$ and $AFmc$ components of the (220) reflection indicate the changing concentration of the two phases, with $Fmc$ dominating near 310 K and $AFmc$ dominating at 80 K. The anisotropic nature of the elastic response to the magnetic phase transitions is illustrated in the schematic (Fig. 6) of the crystal planes involved in the two reflections shown in Fig. 5. The substantial increase of the (220) peak angle in the $AFmc$ phase indicates drastic, uniform contraction of the $a$-$b$ plane, coupled with a mild contraction along the $c$-axis. Equally the broadness of the (220) reflection indicates large local atomic displacements in the $a$-$b$ plane, just as the relative narrowness of the (008) reflection indicates rather minor local atomic displacements in the direction of the $c$ axis.

Close examination of the high resolution X-ray diffraction patterns of the $PrMn_2Ge_{2-x}Si_x$, series collected at 450 K reveals a separation into two distinct phases in all of the pseudo-ternaries. Since this is seen in the paramagnetic phase it cannot be related to the magnetic order and must therefore be chemical in nature. Rietveld analysis of these diffraction patterns provides further insights. These are summarized in Figs. 7(a)–7(c), where the two phases are denoted as Si rich (with blue symbols) and Ge rich (with orange symbols).

Both $a$ and $c$ lattice parameters and the Si/Ge concentration were fitted in the Rietveld refinement process. The concentration dependence of the lattice parameters (Fig. 7(a)) shows a displacement to the left for the Ge rich phase and to the right for the Si rich phase. Although the slope of the concentration dependence of lattice parameters for the two phases is not identical they are quite close to each other, and to the value predicted by Vegard’s law (indicated by full and dashed lines in Fig. 7(a)). The relative concentration of the two phases (shown in Fig. 7(b)) changes sharply for $x \leq 0.8$, but then appears to be relatively constant at ~75% of the Ge rich phase and ~25% of the Si rich phase for all higher concentrations of Si. This rather surprising result indicates that the difference between Si/Ge concentration of the two phases remains relatively constant across the series. Finally, the lattice strain (shown in Fig. 7(c)) is high even in the end compounds ($PrMn_2Ge_2$ and $PrMn_2Si_2$) and is greater in the Ge rich phase than the Si rich phase, increasing to quite large values around $x = 1$.

D. Effect of mechanical pressure

Thus, far we have considered the effect of chemical distributions on the magnetic and structural properties. We now consider the effect of mechanical pressure.

Fig. 8 shows the measured pressure dependence of lattice volume (Fig. 8(a)) and $c/a$ ratio (Fig. 8(b)) for three compounds in this series. The data were obtained by X-ray diffraction on the PD beamline at the Australian Synchrotron, using a diamond anvil pressure cell. The compressibility of $PrMn_2Ge_2$ is noticeably greater than $Pr_{0.5}Y_{0.5}Mn_2Ge_2$ and $PrMn_2Si_2$, particularly at lower pressures. We note that both $PrMn_2Ge_2$ and $Pr_{0.5}Y_{0.5}Mn_2Ge_2$ adopt the $Fmc$ state at
ambient temperature and pressure, whereas PrMn$_2$Si$_2$ is in the AFmc state.

We also note from the report of pressure dependence of magnetization in Pr$_{0.5}$Y$_{0.5}$Mn$_2$Ge$_2$ (Ref. 3) that the critical pressure for the Fmc $\rightarrow$ AFmc transition occurs at P/C0 6.7 kilobars, as indicated by the grey cross in Fig. 8(a). Taking this as a general indication of critical volume for the Fmc $\rightarrow$ AFmc transition, we deduce that the critical pressure for the transition in PrMn$_2$Ge$_2$ lies at P/C0 38 kilobars.

This interpretation is well supported by the trends in c/a ratios (Fig. 8(b)), where we see a pronounced increase in the c/a ratio of Pr$_{0.5}$Y$_{0.5}$Mn$_2$Ge$_2$ from very low pressure and an increase in slope of the c/a ratio for PrMn$_2$Ge$_2$ at P = 38 kilobars. In contrast, the c/a ratio of PrMn$_2$Si$_2$, which is in the AFmc state under ambient conditions, does not increase with pressure.

E. Comparison of chemical and mechanical pressure

In order to compare the effects of mechanical pressure with those of chemical distributions, we describe the lattice response to pressure with the equation of state whereby, the bulk modulus ($B$) describes the change in volume ($V$) of a solid in response to pressure ($P$)

$$B = -V \left( \frac{\delta P}{\delta V} \right)_T. $$ (1)

Defining ($B'$) to describe the pressure dependence of the bulk modulus, such that

$$B' = \left( \frac{\delta B}{\delta P} \right)_T. $$ (2)

In practice $B'$ is generally constant and can be reduced to $B'_o$, so we can write

$$B = B_o + B'_o P, $$ (3)

where $B_o$ is $B$ at ambient pressure, and we can integrate Eq. (1) to give

$$P(V) = B_o \left( \frac{V_o}{V} \right)^{B'_o} - 1), $$ (4a)

or equivalently

$$V(P) = V_o \left( 1 + B'_o \frac{P}{B_o} \right)^{-1/B'_o}, $$ (4b)

where $V_o$ is the volume of the solid at ambient pressure.

We have applied Eq. (4a) to our room temperature diffraction data with values of $B_o$ and $B'_o$ from a closely related compound (UGe$_2$Mn$_2$) (Ref. 13) to derive the equivalent “chemical pressure” from our three series of pseudo-ternary compounds.

The results are summarized in Fig. 9, where we plot chemical pressure against substituent concentration ($x$). The dashed lines in Fig. 9 broadly indicate the slope of chemical pressure for each compound, and highlight the transition at $x = 0.5$, which marks the boundary between Fmc and AFmc magnetic state in Pr$_{1-x}$Y$_x$Mn$_2$Ge$_2$ (Ref. 3) and PrMn$_2$(Ge$_{1-x}$Si$_x$)$_2$ (Ref. 2).

For those compounds, we see a marked increase in compressibility in the AFmc phase. In contrast, Pr(Mn$_{1-x}$Fe$_x$)$_2$Ge$_2$
undergoes a magnetic transition from $Fmc$ to $AFI$ near $x = 0.1$ and another to $PM$ near $x = 0.7$. Clearly, neither magnetic transition affects the compressibility of the Pr$(Mn_{1-x}Fe_x)2Ge_2$ compound.

The initial slope of the change in chemical pressure with concentration reflects the magnitude of response to the mismatch between atom sizes in the mixed lattice plane, in the absence of any magnetic transition. For Pr$_{1-x}$Y$_x$Mn$_2$Ge$_2$ and Pr$_{1-x}$Mn$_2$(Ge$_{1-x}$Si$_x$)$_2$, the initial slope is roughly in proportion to the relative sizes of the ions in the mixed plane (i.e., Y$^{3+}$ is $\approx 10\%$ smaller than Pr$^{3+}$ and Si$^{4+}$ is $\approx 25\%$ smaller than Ge$^{4+}$). It is perhaps surprising that substitution of Fe for Mn, where the electron density distribution is only slightly more compact, applies by far the strongest chemical pressure. It may be that dilution of the Mn-Mn exchange interaction with increasing Fe concentration produces an effective increase in compressibility.

Finally, we compare the relative efficiencies of mechanical and chemical pressure on PrMn$_2$Ge$_2$. Although Fig. 9 indicates that the magnetic $Fmc \leftrightarrow AFmc$ transition in Pr$_{1-x}$Y$_x$Mn$_2$Ge$_2$ and Pr$_{1-x}$Mn$_2$(Ge$_{1-x}$Si$_x$)$_2$ occurs around $x = 0.5$, we note that both Pr$_{0.5}$Y$_{0.5}$Mn$_2$Ge$_2$ and Pr$_{0.5}$GeSi actually display mixed $Fmc/AFmc$ character at room temperature, and the boundary is not sharp. The chemical boundary for complete conversion to the AFmc magnetic state at room temperature is closer to Pr$_{0.4}$Y$_{0.6}$Mn$_2$Ge$_2$ (see Fig. 3) and PrMn$_2$Ge$_{0.6}$Si$_{1.4}$ (Ref. 2), corresponding to chemical pressure of $P \approx 27$ kilobars (for Y substitution) and 55 kilobars (for Si substitution). These pressures are comparable to our critical value of $P \approx 38$ kilobars for applied mechanical pressure. Given the coarseness of our experimental data (in terms of chemical substitution), this result is in good agreement with our comparison of the rate of change of Pr ordering temperature ($T_{Cm}^{Pr}$), which indicates that mechanical pressure is a more efficient driver of the $Fmc$ to $AFmc$ magnetic transition than chemical pressure. If indeed chemical pressure is less effective, it may derive from the enhanced local strain distribution and phase separation which accompanies chemical substitution.

IV. CONCLUSIONS

We have shown that the two phase region in the PrMn$_2$Ge$_2$-$x$Si$_x$, Pr$_{1-x}$Y$_x$Mn$_2$Ge$_2$, and PrMn$_2$-$x$Fe$_x$Ge$_2$ series of compounds is associated with chemical phase separation due to chemical distributions at the mixed site. We have also shown that chemical substitution produces large local strain distributions, mainly in the $a$-$b$ planes. Finally, we have shown that these compounds all share a common magnetic character, dependent mainly on the Mn-Mn interatomic distance, regardless of which site bears the substituent atoms, and that this character is broadly consistent with the effect of mechanical pressure.

ACKNOWLEDGMENTS

We wish to thank Andrew Studer (ANSTO), Zhengxiang Cheng and Muhammad Faiz Md Din (ISEM, Wollongong), Luana Caron and Ekkes Brück (TU-Delft), and Qinfen Gu and Helen Brand (Australian Synchrotron), for their valuable contributions to this research.

We also acknowledge the Australian Research Council (ARC), the Australian Institute of Nuclear Science and Engineering (AINSE), and the Access to Major Research Facilities Program (AMRFIP) for their sustained financial support.