Theoretical calculation of magnetic structure variation in Pr5Ni2Si3

S. H. Song, J. E. Snyder, and D. C. Jiles

Citation: J. Appl. Phys. 99, 08P304 (2006); doi: 10.1063/1.2173608
View online: http://dx.doi.org/10.1063/1.2173608
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v99/i8
Published by the American Institute of Physics.

Theoretical calculation of magnetic structure variation in $Pr_5Ni_2Si_3$

S. H. Song,[a] J. E. Snyder, and D. C. Jiles
Materials and Engineering Physics Program, Ames Laboratory, U.S. Department of Energy, and Materials Science and Engineering Department, Iowa State University, Ames, Iowa 50011

(Presented on 2 November 2005; published online 27 April 2006)

The variation of magnetization with temperature of the $Pr_5Ni_2Si_3$ compound was calculated using a nearest neighbor exchange interaction approximation. Pr atoms, which are the only element in this compound with a net magnetic moment, were classified into three types based on the number of nearest neighbor exchange interactions. The expected magnetization versus temperature curve for each type of Pr atom was calculated using the Brillouin function, as well as the average magnetization versus temperature curve for the entire unit cell. The results show that the "corner" atoms exhibit very different behavior from that of the other types of Pr atoms on the "center" or "edge" sites. This is due to the broken symmetry in exchange interaction at the corner site due to interactions with atoms from outside the unit cell that are in closer proximity than atoms within the unit cell. This is considered to be the cause of a second magnetic phase transition observed at a lower temperature than the Curie temperature. © *2006 American Institute of Physics*.
[DOI: 10.1063/1.2173608]

A "model system" in materials science needs to satisfy at least three conditions. (1) The system must be simple enough that there are only a few key dominant factors which influence the physical property to be studied or predicted. (2) The "control variables" that are ultimately responsible for the physical phenomena (such as composition, crystal structure, temperature, pressure, or exchange interaction) should be alterable in a well defined way so that their effects can be studied independently. (3) The general ideas and predictions obtained from the model system should be applicable to other related cases. In this respect, the homologous series of Pr–Ni–Si compounds can serve as a model system for the study of structure/property relationships in magnetic materials.

The ternary Pr–Ni–Si system contains the homologous series of compounds $R_{(n+2)(n+1)}Ni_{n(n-1)+2}Si_{n(n+1)}$, where R is a rare earth element, in this case Pr. It provides a range of materials with different structures as described by Rogl.[1] The compounds $Pr_6Ni_2Si_3(n=2)$, $Pr_5Ni_2Si_3(n=3)$, and $Pr_{15}Ni_7Si_{10}(n=4)$ have been prepared and are known to be the members of this family. These compounds exhibit a hexagonal structure formed of trigonal prismatic columns in which the size of the base plane of the trigonal prismatic "cluster" is determined by the value of n in the chemical formula. As the size of the cluster changes, the number of exchange interactions for each Pr atom systematically changes, thus the physical properties are expected to vary systematically from one member of the series to the next.

Experimental investigations of $Pr_5Ni_2Si_3$ and $Pr_{15}Ni_7Si_{10}$ polycrystalline samples and a $Pr_5Ni_2Si_3$ single crystal sample have recently been reported.[2–5] According to the experimental results, each compound shows two magnetic phase transitions: a magnetic order/disorder transition at a higher temperature (41 K for $Pr_5Ni_2Si_3$ and 58 K for $Pr_{15}Ni_7Si_{10}$) and another transition, which exhibits characteristics of spin reorientation transition, at a lower temperature (25 K for $Pr_5Ni_2Si_3$ and 31 K for $Pr_{15}Ni_7Si_{10}$). The present paper reports on the theoretical investigation of temperature dependent magnetization of single crystal $Pr_5Ni_2Si_3$. The calculations are based on a nearest neighbor exchange interaction approximation. Studies of next nearest neighbor interactions would also be of interest. The inclusion of such additional interactions will require a reduction of the pair exchange energy to maintain the same Curie temperature, and should reduce the sharpness of the Curie point transition compared with the calculation based only on nearest neighbors.

The exchange interaction energy for each atomic site was determined using a nearest neighbor exchange interaction approximation. For this calculation, it was assumed that (1) only the Pr atoms contribute to magnetization and for the purposes of the calculation the magnetic moment per Pr atom was assumed to be $3.58\mu_B$, which is the theoretical value for an isolated Pr atom. In practice the moment on the Pr in condensed matter is smaller, and the Pr atoms on the different lattice sites also have different values of magnetic moment. Therefore for an attempt at such a calculation a fixed magnitude of moment was assumed. (2) The exchange interaction exists only between the nearest neighbors, which is expressed as

$$E_{ex} = -2J_{NN}\sum_{i,j} J_i \cdot J_j,$$

where J_{NN} is the exchange interaction constant between nearest neighbors and J_i and J_j are total angular momentum at the ith and jth sites, respectively.

The Pr atoms in the triangular base plane of the structure of $Pr_5Ni_2Si_3$ [Figs. 1(a) and 1(b)] can be classified into three groups by the number of nearest neighbors among Pr atoms: Pr atoms at the center sites, along the edges, and at the corners of the triangular plane have eight, ten, and eleven nearest neighbors, respectively [Fig. 1(b)]. The average exchange

[a]Electronic mail: song73@iastate.edu

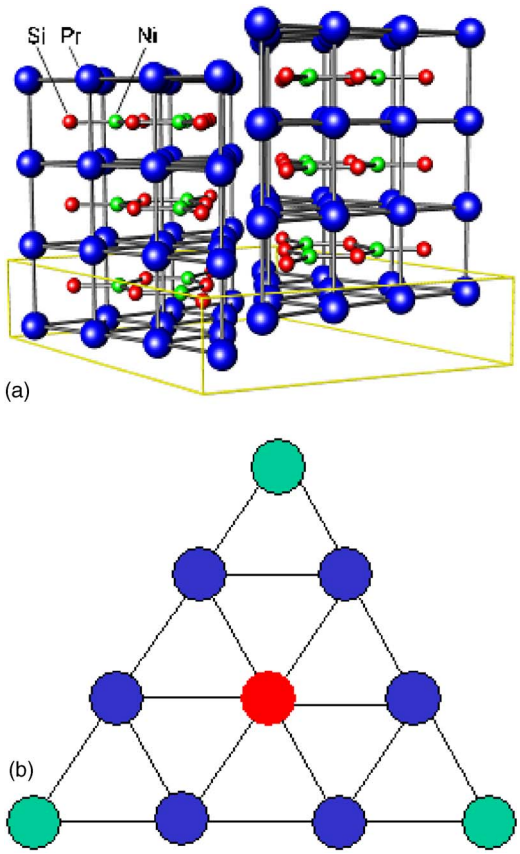

FIG. 1. (a) Crystal structure and unit cell of $Pr_5Ni_2Si_3$ showing the trigonal cells which form prismatic columnar assemblies. [Note that for the purposes of clarity there are additional Ni atoms not shown here which lie at the corner of the rhombohedra containing the two trigonal prisms and amount to one extra Ni atom per trigonal plane (Ref. 5)]. (b) Schematic picture describing three types of Pr atoms in the base plane of pristimatic columnar structure of $Pr_5Ni_2Si_3$ single crystal.

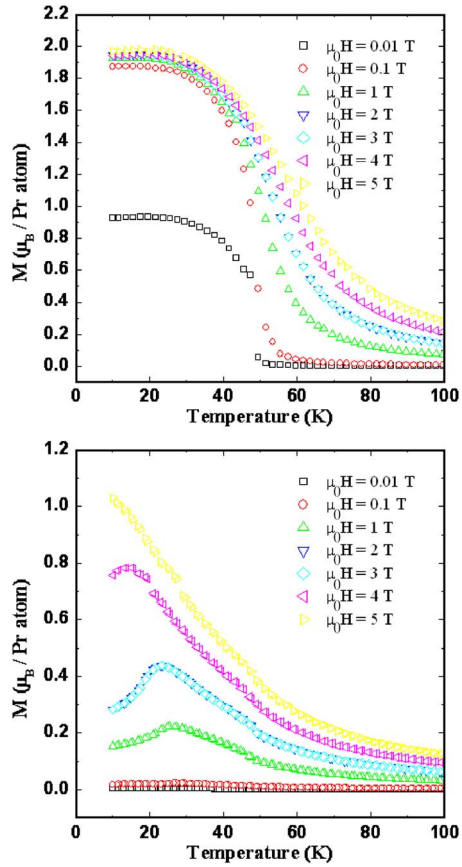

FIG. 2. Variation of the magnetization of $Pr_5Ni_2Si_3$ single crystal with temperature, measured by SQUID (a) parallel and (b) perpendicular to c axis under various magnetic fields (Ref. 6).

interaction energy between nearest neighbors was calculated based on the experimentally measured Curie temperature (T_c).

Since the effective magnetic field that each atom experiences depends on the number of nearest neighbors for each site, the effective magnetic field for each atomic site is determined from

$$E_{\mathrm{ex}}^{\mathrm{site}} = -J_{\mathrm{NN}} \sum_{i,j}^{\mathrm{site}} J_i \cdot J_j = m \cdot \mu_0 H_{\mathrm{eff}}^{\mathrm{site}} = m \cdot \mu_0 \alpha_{\mathrm{site}} M,$$

where $H_{\mathrm{eff}}^{\mathrm{site}}$ is the molecular field and α_{site} is the molecular field constant for that site.

The total magnetization can be expressed as the sum of the magnetic moments of the three different types of atoms,

$$m^{\mathrm{triangle}} = m^c + m^e + m^a = m_0 B_J \left[\frac{m_0 \mu_0 (H + H_{\mathrm{eff}}^c)}{k_B T} \right] + 6 m_0 B_J \left[\frac{m_0 \mu_0 (H + H_{\mathrm{eff}}^e)}{k_B T} \right] + 3 m_0 B_J \left[\frac{m_0 \mu_0 (H + H_{\mathrm{eff}}^a)}{k_B T} \right],$$

where B_J is the Brillouin function and m^c, m^e, and m^a are the magnetization due to each of the Pr atoms at the center, edge, and corner of the triangular plane, respectively.

Figures 2(a) and 2(b) show M vs T curves of $Pr_5Ni_2Si_3$ single crystal, where M was measured parallel (a) and perpendicular (b) to the c axis using a superconducting quantum interference device (SQUID) magnetometer.[6] The SQUID measurements appear to show that the magnetization is not saturated in either direction even at a field of $\mu_0 H = 5$ T but that the easy axis appears tilted much closer to the c axis than to the base plane. This can be explained by considering the atomic arrangement of Pr atoms. For a Pr atom at the corner site, the Pr atom has 11 nearest neighbor Pr atoms, as shown in Fig. 3. This indicates that the sixfold symmetry in exchange interaction is broken by the lack of one Pr atom at one of the six nearest neighbor sites in the same plane. As a result the magnetic moments at the corner Pr atoms will tend to be tilted away from the c axis, as confirmed by the neutron diffraction data[7] which indicate that the magnetic moment of the Pr atom at the corner site is $0.72\mu_B$ along the c axis. Considering that the theoretical magnetic moment of Pr atom is $3.58\mu_B$, the measured magnitude of $0.72\mu_B$ along the c axis can be interpreted as the magnetic moment of the corner atoms is tilted 78° away from the c axis. Based on these results, the angles between the magnetic moments for each site were calculated and these were included in the calculation of the exchange interaction energy,

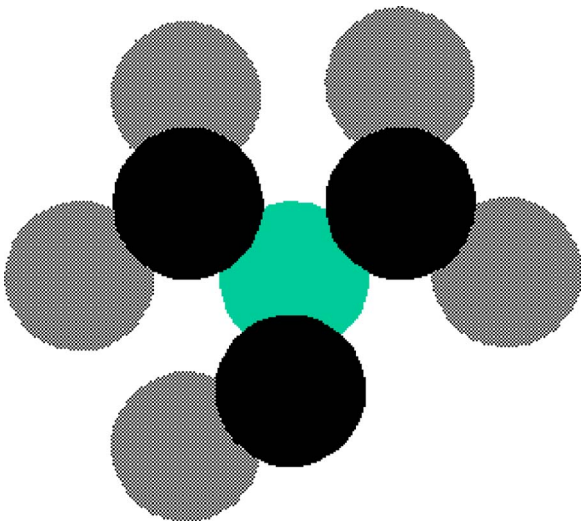

FIG. 3. Atomic arrangement of Pr atoms around the corner site (looking along the c axis). The central sphere is a Pr atom at the corner site which has 11 nearest neighbor Pr atoms: 5 in the same base plane, 2 in the same trigonal column and 3 in the second neighbor columns (grey spheres); 3 in neighboring trigonal columns 1/2 plane up (black spheres); and 3 in neighboring trigonal columns 1/2 plane down (not visible behind the black spheres).

$$E_{\mathrm{ex}} = -2J_{\mathrm{NN}} \sum_{i,j} J_i \cdot J_j \cos\,\theta,$$

where θ is the angle between magnetic moments at ith and jth sites.

Based on the calculated exchange energy the expected M vs T curves for each type of Pr atom were calculated. The results are shown in Fig. 4, which shows a consistent Curie temperature for all sites.

The measured $M_{\perp c}$-T curve [Fig. 2(b)] exhibits a local maximum in magnetization at 25 K under an applied magnetic field of 1 T and the transition temperature where the

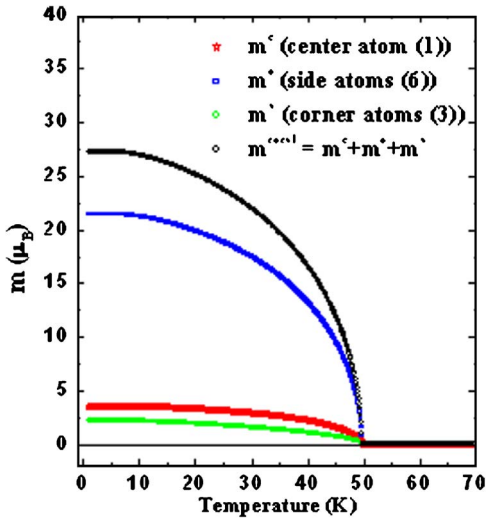

FIG. 4. Simulated M vs T curves using Brillouin functions for the $Pr_5Ni_2Si_3$ compound. Red, blue, and green symbols represent the magnetization due to the Pr atoms at the center, edge, and corner sites, respectively. The black symbols are the sum of these three contributions, which is the total magnetic moment of the ten Pr atoms in the triangular plane.

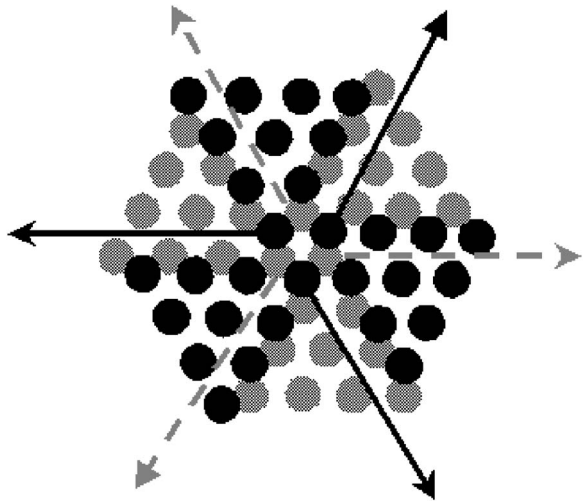

FIG. 5. Atomic arrangement of Pr atoms. Each arrow from the corner atoms indicates the direction in which the symmetry of the exchange interaction at the corner atom is broken.

local maximum occurs is lowered as the magnitude of field increases. This behavior is also considered to be due to the asymmetric exchange interactions at the corner sites. The direction of the magnetic component of the corner atom in the base plane should tend to be along the direction of broken symmetry, which is indicated with arrows in Fig. 5, because thermal agitation encounters no competing influence from exchange interaction in this direction, thus making it easy for the magnetic moment to tilt to this direction. Therefore the magnetic components of the corner atoms on the base plane tend to turn to their own locally preferred directions (arrows in Fig. 5). As a consequence, when the temperature decreases below a critical temperature the total magnetic moments on the base plane will be compensated, meaning that the moments of the corner atoms are aligned in such a way that they partially cancel each other out, thus reducing the component of magnetization in the plane.

This research was supported by the U.S. Department of Energy, Office of Science (OS), Office of Basic Energy Sciences (BES), Materials Sciences Division. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-ENG-82. The authors wish to thank Dr. R. W. McCallum for the access to unpublished single crystal $Pr_5Ni_2Si_3$ measurements and for helpful discussions concerning the results of this research.

[1] P. Rogl, in *Handbook of the Physics and Chemistry of Rare Earths*, edited by K. A. Gschneidner, Jr. and L. R. Eyring (North-Holland, Amsterdam, 1987), Vol. 7, p. 1.
[2] A. O. Pecharsky, Y. Mozharivskyj, K. W. Dennis, K. A. Gschneidner, R. W. McCallum, G. J. Miller, and V. K. Pecharsky, Phys. Rev. B **68**, 134452 (2003).
[3] S. H. Song, D. C. Jiles, J. E. Snyder, A. O. Pecharsky, D. Wu, K. W. Dennis, T. A. Lograsso, and R. W. McCallum, J. Appl. Phys. **97**, 10M516-1 (2005).
[4] S. H. Song, J. E. Snyder, D. Wu, T. A. Lograsso, K. W. Dennis, R. W. McCallum, Y. Janssen, and D. C. Jiles, IEEE Trans. Magn. **41**, 3499 (2005).
[5] D. C. Jiles *et al.*, J. Magn. Magn. Mater. **299**, 288 (2006).
[6] R. W. McCallum (private communication).
[7] A. Llobet Megias (private communication).