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Theoretical calculation of magnetic structure variation in Pr$_5$Ni$_2$Si$_3$

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

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The variation of magnetization with temperature of the Pr$_5$Ni$_2$Si$_3$ 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.

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$_n$Si$_{n+1}$ (where R is a rare earth element, in this case Pr). It provides a range of materials with different structures as described by Rogl. The compounds Pr$_5$Ni$_2$Si$_3$ (n = 2), Pr$_5$Ni$_2$Si$_3$ (n = 3), and Pr$_5$Ni$_2$Si$_3$ (n = 4) have prepared and are known to 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$_5$Ni$_2$Si$_3$ and Pr$_5$Ni$_2$Si$_3$ polycrystalline samples and a Pr$_5$Ni$_2$Si$_3$ single crystal sample have been reported. Accoring to the experimental results, each compound shows two magnetic phase transitions: a magnetic order/disorder transition at a higher temperature (41 K for Pr$_5$Ni$_2$Si$_3$ and 58 K for Pr$_5$Ni$_2$Si$_3$) and another transition, which exhibits characteristics of spin reorientation transition, at a lower temperature (25 K for Pr$_5$Ni$_2$Si$_3$ and 31 K for Pr$_5$Ni$_2$Si$_3$). The present paper reports on the theoretical investigation of temperature dependent magnetization of single crystal Pr$_5$Ni$_2$Si$_3$. 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 magnetic 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$_5$Ni$_2$Si$_3$ [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

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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

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

where $H_{eff}^{site}$ is the molecular field and $\alpha_{site}$ 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,

$$m^{\text{triangle}} = m^c + m^e + m^a = m_0 B_f \left[ \frac{m_0 \mu_0 (H + H_{eff}^c)}{k_B T} \right]$$

$$+ 6m_0 B_f \left[ \frac{m_0 \mu_0 (H + H_{eff}^e)}{k_B T} \right]$$

$$+ 3m_0 B_f \left[ \frac{m_0 \mu_0 (H + H_{eff}^a)}{k_B T} \right],$$

where $B_f$ 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$_5$Ni$_2$Si$_3$ 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 $H = 5$ 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.
\[ E_{\text{ex}} = -2J_{\text{NN}} \sum_{i,j} J_i \cdot J_j \cos \theta, \]

where \( \theta \) is the angle between magnetic moments at \( i \)th and \( j \)th 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 \( M \) vs \( T \) 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 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.

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6R. W. McCallum (private communication).
7A. Llobet Megias (private communication).