Magnetic properties of Pm in NdNi

K. Nishimura and K. Mori
Faculty of Engineering, Toyama University, Toyama 930, Japan

S. Ohya
Faculty of Science, Niigata University, Niigata 950-21, Japan

S. Muto
National Laboratory for High Energy Physics, Tsukuba 305, Japan

Y. Isikawa
Faculty of Science, Toyama University, Toyama 930, Japan

(Received 4 August 1995; revised manuscript received 2 November 1995)

Magnetic properties of Pm as an impurity in NdNi single crystal were investigated by means of low-temperature nuclear orientation of the ^{144}Pm isotope. The angular distribution of γ-ray anisotropy revealed that the direction of the hyperfine field experienced by the nuclei was in the (a,c) plane and made an angle of 29(10)° and/or 209(10)° with the a axis. The strength of the field was deduced to be 395(48) T with a full-field site fraction of 76(3)% from the temperature dependence of the anisotropy. The β decay of ^{144}Pm was found to proceed mainly via the $\Delta J_\beta = 1$ matrix element. A brief discussion of crystal electric field effects and exchange interactions is given. [S0163-1829(96)01722-5]

I. INTRODUCTION

Rare-earth intermetallic compounds have received considerable interest concerning magnetic properties arising from 4 f-electron motion. RNi (R = light rare-earth elements) is one of those systems which show a large magnetic anisotropy due to the crystal electric-field (CEF) effect. A number of studies have been carried out to investigate the magnetic structures.[1–4] However, very little is known about PmNi because of the lack of a stable Pm isotope. This work aims to offer information on the magnetic structure and hyperfine field (HF) of the Pm ions in a NdNi host compound by the technique of low-temperature nuclear orientation (LTNO). When the nuclear properties of probe radioactive nuclei are well known, this technique can determine the strength and direction of the HF acting at the nuclei under study. These in turn relate to the magnitude of the electronic magnetic moments of the oriented ions and their spatial ordering. Particularly for rare-earth ions, in which the main contribution to the HF arises from orbital motion of the localized $4f$ electrons, the magnetic structure can be investigated through such hyperfine interaction studies. The host compound of NdNi is ferromagnetic, and the magnetic structure has been determined.[3] The present result is briefly discussed together with that of our previous work for the Pm ions in PrNi.[5]

II. EXPERIMENT

The crystal structure of NdNi is orthorhombic of CrB type (space group $Cmcm$).[6] A single crystal of NdNi was prepared by the Czochralski pulling method in an argon-gas atmosphere. The crystal was cut into a rectangular plate of dimensions: $3\times0.2\times2$ mm along the a, b, and c axes, respectively. A cyclotron irradiated the sample with a 1 μA deuteron beam for 30 min at the Institute for Nuclear Study, University of Tokyo. By the reactions ^{143}Nd(d,n) and ^{144}Nd$(d,2n)$, the activity of ^{144}Pm ($T_{1/2}=349$ d) was produced in the sample about 9 μCi. During the irradiation some other activities, such as ^{143}Pm and ^{148}Pm, were also produced. The total concentration ratio of Pm to Nd is approximately 1 ppm. The sample was kept in vacuum for half a year to allow decays of short-life activities before being soft-soldered to a copper cold finger of a ^{3}He/^{4}He dilution refrigerator. A ^{60}Co in Co(hcp) nuclear thermometer was also soldered near the sample to monitor the temperature. NdNi is easily magnetized in the direction of 23.5° with the a axis in the (a,c) plane.[3] We set the easy magnetization direction and the c axis of Co(hcp) parallel, the (a,c) plane of NdNi being horizontally or vertically, so that the γ-ray angular distribution of ^{60}Co nuclei showed spatial directions of the crystallographic axes of NdNi. Two pairs of Helmholtz-type coils, i.e., the four coils being at right angles to each other, produced an external field of 0.4 T horizontally along the easy magnetization direction. Four Ge detectors were set on a goniometer to measure γ-ray energy spectra.

III. RESULT

The γ-ray anisotropy $W(\Theta)$ was obtained from the cold-counting rate divided by the warm rate, where 4.2 K is considered warm enough to ensure isotropic radiation. Interpretation of orientation results has been made using the expression[7]

$$W(\Theta)=1+f\sum_{\lambda} B_\lambda(\mu,B_{\rm HF},T)U_\lambda(\Delta J_\beta)A_\lambda(L_\gamma)\times Q_\lambda P_\lambda(\cos\Theta),$$

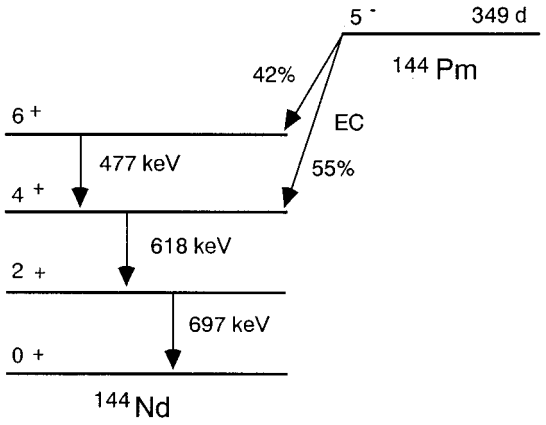

FIG. 1. Simplified decay scheme of ^{144}Pm.

where Θ specifies an angle between the orientation axis and the emission direction of radiation. The factor f denotes the fraction of nuclei which experience the fullfield site, the remainder $(1-f)$ being taken to experience zero field.[8] The summation index λ is limited to 2 and 4 when a deexcitation γ transition has a pure $E2$ character. The orientation parameters B_λ describe the degree of orientation, depending on the nuclear magnetic moment, the strength of the HF and temperature. U_λ and A_λ depend only on the decay mode of the isotope under study: the nuclear spin I, angular momentum of the lepton pair ΔJ_β, and the γ radiation multipolarity L_γ. A simplified decay scheme is shown in Fig. 1.[9] The β decay is first-forbidden of $|\Delta I|=1$, so that the U_λ could have two terms corresponding to the cases where the lepton pair carries unit angular momentum ($\Delta J_\beta=1$) or two angular momentum ($\Delta J_\beta=2$). The fractional contribution of each term will be deduced from temperature dependences of γ anisotropy as described below. The γ deexcitations are most likely $E2$ transitions, i.e., $L_\gamma=2$. The Q_λ are the solid-angle correction factors of the detectors used; $Q_2=0.98$ and $Q_4=0.95$. The Legendre polynomials P_λ are evaluated at the angle each detector subtends at the orientation axis.

Figure 2(a) shows the γ anisotropy as a function of angle θ measured from the a axis in the (a,c) plane at the base temperature of 8.5 mK. It is obvious that the nuclear magnetic moment was oriented at an angle of 29(10)° with respect to the a axis. The HF produced by the $4f$ electrons of Pm ions are in the same direction. The anisotropy between the 29° direction and the b axis also showed that the orientation axis was in the 29° direction in the (a,c) plane as shown in Fig. 2(b). We cannot rule out the case in which the nuclei oriented at 29° and/or 209° because the γ anisotropy does not distinguish between ferro- and antiferromagnetic orders.

As shown in Fig. 3 the temperature dependences of the anisotropy were measured at angles of 29° and 119°, parallel and perpendicular to the orientation axis, respectively. The data were analyzed to extract the hyperfine interaction strength, ratios of U_4/U_2 in the β decay[10] and the full-field site fraction f. The hyperfine interaction strength was deduced to be $3.37(13)\times10^{-24}$ J. Taking the nuclear magnetic moment of ^{144}Pm to be $1.69(14)\mu_N$,[9] the hyperfine field was obtained as $B_{\rm HF}=395(48)$ T.

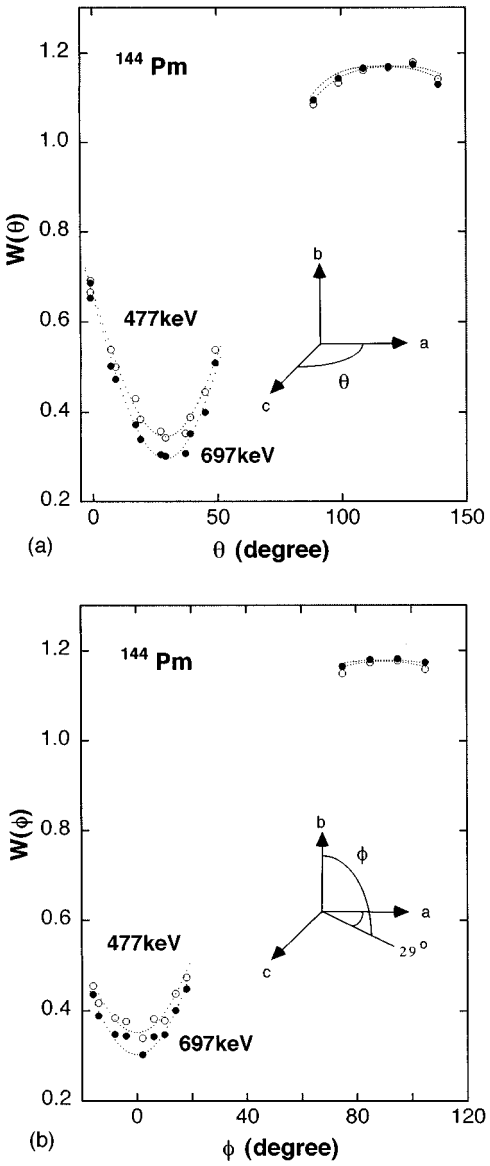

FIG. 2. Angular dependence of γ-ray anisotropy of ^{144}Pm in the (a,c) plane. (b) Angular dependence of γ-ray anisotropy of ^{144}Pm between the b axis and the orientation axis.

Concerning the U_λ parameters, one should consider a mixing of the β-decay matrix elements for $\Delta J_\beta=1$ and 2. Letting δ^2 be the ratio of $U_\lambda(\Delta J_\beta=1)$ and $U_\lambda(\Delta J_\beta=2)$, the U_4/U_2 ratio can be given as[7]

$$U_4/U_2=\frac{U_4(1)+\delta^2U_4(2)}{U_2(1)+\delta^2U_2(2)}.$$

The fit for the 477 keV transition data, taking fU_2 and fU_4 as parameters, yielded a value $U_4/U_2=0.915(25)$ which results in $\delta^2=0.0(+0.02)$. It means that the β-decay matrix element of $\Delta J_\beta=1$ is dominant in $5^-\rightarrow6^+$. Assuming $\delta^2=0$ for the 477 keV transition data, we estimated the ratio in the other branch, $5^-\rightarrow4^+$, using the 697 keV transition data. The fit gave a value of $U_4/U_2=0.290(7)$, which leads to $\delta^2=0.07(^{-0.07}_{+0.08})$. The 618 keV photon peak accidentally overlapped with another one from ^{148}Pm, so we could not extract the ratio from this tran-

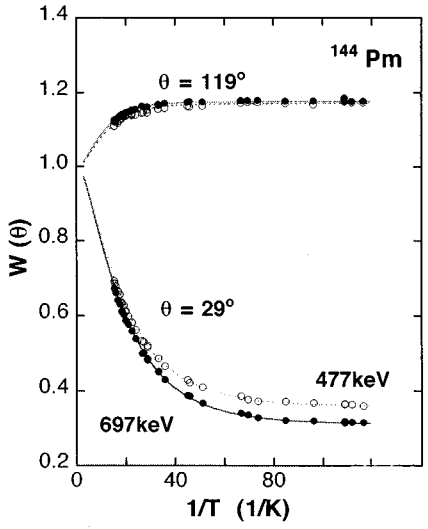

FIG. 3. Temperature dependence of γ-ray anisotropy of ^{144}Pm in the (a,c) plane.

sition data. The fraction of nuclei in the full-field site was derived as $f=0.76(3)$ from the above analysis.

IV. DISCUSSION

In the previous study of Pm in PrNi, the orientation axis was found to make an angle of 30(5)° with the a axis in the (a,c) plane.[5] The present work shows that Pm nuclei in NdNi orients also at 29(10)° with the a axis in the (a,c) plane. In brief, the Pm ions have almost the same electronic moment arrangement (magnetic arrangement) in NdNi and PrNi in spite of the different magnetic structure of the host compounds.

The known magnetic structures of PrNi, NdNi, and SmNi are illustrated in Fig. 4.[3,4] PrNi is ferromagnetic below 20 K, the easy magnetization direction being the c axis. The easy magnetization direction of NdNi is the a axis at 28 K, and then turns through 23.5° towards the c axis in the (a,c) plane below 15 K. SmNi has the same crystal structure as those of NdNi and PrNi, but the easy magnetization direction is the b axis below 45 K. Each magnetic structure resembles that of an RGa compound (R=Pr, Nd, or Sm), respectively, which also has the orthorhombic CrB-type crystal structure.[11] In CrB-type crystals, the b axis is a peculiar direction because the lattice constant of the b axis is about twice as large as those of the a and c axes. This asymmetric

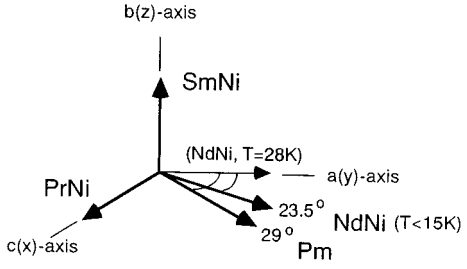

FIG. 4. Magnetic structure of PrNi, NdNi, and SmNi.

crystal property leads to the large magnetic anisotropy observed between the b axis and the b plane due to the CEF effect.

The easy magnetization directions change from the (a,c) plane (the b plane) to the b axis between the Nd and Sm compounds. As described in the literature,[11,12] such changes in magnetic structures correspond to the sign change in the second-order Stevens coefficient[13] α_J between Nd^{3+} and Sm^{3+}; the α_J values for Pr^{3+}, Nd^{3+}, and Sm^{3+} are -2.1×10^{-2}, -0.64×10^{-2}, and $+4.1\times10^{-2}$, respectively. By use of the Stevens operator equivalents O_l^m and the CEF parameters V_l^m, the lowest-order terms of the CEF Hamiltonian can be expressed as[14]

$$H_{\mathrm{CEF}}=\alpha_J(V_2^0O_2^0+V_2^2O_2^2)$$
$$=\alpha_JV_2^0[3J_z^2-J(J+1)]+\alpha_JV_2^2(J_x^2-J_y^2).$$

Assigning x, y, and z axes to the crystallographic c, a, and b axes, respectively, a simple point-charge model predicts the values $V_2^0\simeq-140$ K and $V_2^2\simeq-60$ K. We assume a 3+charge for rare-earth ions and zero charge for Ni in the calculation. These V_l^m values were found not to change very much between PrNi, NdNi, and SmNi. The calculations explain the change in the easy magnetic directions between NdNi and SmNi. The off-axis ordering of NdNi is probably accounted for by including fourth-order terms of the CEF Hamiltonian. In order to understand the c-axis ordering of PrNi, a positive value of V_2^2 is necessary, so the present calculation is not valid for this compound. Positive V_2^2 could be obtained by varying the crystallographic parameters.[3]

Following the above argument, the electronic magnetic moments of Pm^{3+} would order along the b axis owing to the positive a_J value of 0.77×10^{-2} as far as only the second-order CEF Hamiltonian is concerned. The observed orientation axes of Pm nuclei in NdNi and PrNi seem to suggest that higher-order terms of the CEF Hamiltonian are important. Furthermore, there exist exchange interactions between Pm and Nd or Pr ions, which are responsible for magnetic ordering. We believe that the magnetic arrangements of Pm ions are made by a balance of the CEF effect and the exchange interactions.

The HF of rare-earth ions can be written as[15]

$$B_{\mathrm{HF}}=B_{4f}+B_{\mathrm{sp}}+B_{NN}+B_{\mathrm{appl}},$$

where B_{4f} depends on the total angular momentum of $4f$ electrons of the ion; $B_{4f}=A\langle J_z\rangle$. The factor A is the hyperfine coupling constant.[15] B_{sp} is due to the conduction electron self-polarization by the $4f$ electrons, B_{NN} is the transferred field through conduction electrons polarized by magnetic neighbor ions, and B_{appl} is an applied field. The experimental and theoretical values of B_{HF} related to this discussion are listed in Table I. $B_{\mathrm{HF}}^{\mathrm{expt}}$ of Pm ions in NdNi is close to $B_{\mathrm{HF}}^{\mathrm{theor}}$. $B_{\mathrm{HF}}^{\mathrm{expt}}$ of Pm ions in PrNi, however, differs greatly from $B_{\mathrm{HF}}^{\mathrm{theor}}$. The reduction of $B_{\mathrm{HF}}^{\mathrm{expt}}$ in PrNi could be attributed to the CEF effect and the exchange interactions. The CEF effect often quenches angular momentum. From

TABLE I. Experimental and theoretical hyperfine fields.

Host	Probe	B_{HF}^{theor} (T) (Ref. 15)	B_{HF}^{expt} (T)	Reference
PrNi	^{142}Pr	337	290(10)	16
NdNi	^{147}Nd	430	418(12)	17
PrNi	^{144}Pm	426	185(22)	5
NdNi	^{144}Pm	426	395(48)	This work

the magnetization measurements of NdNi and PrNi,[3] the values $\langle J_z \rangle \simeq 3.65$ and 2.94 can be deduced, respectively. These values suggest that the angular momentum of Nd and Pr ions are quenched to about 81 and 73 % of the free-ion values of Nd^{3+} and Pr^{3+}, respectively. The angular momentum of Pm ions is probably partially quenched by the CEF effect. This quenching effect on the Pm ions may be larger in PrNi than in NdNi, and thus B_{4f} is perhaps smaller in PrNi.

B_{sp} and B_{NN} are known to make contact interactions with the unbalanced spin density at the nuclear site.[15] There are conduction electrons polarized by the magnetic neighbor ions. Through those electrons, the exchange interactions depends on the angular momentum J of the $4f$ electrons and the conduction electron spin s; $H_{ex} \propto Js$. Since the spatially ordered electronic moments of Pm and Nd ions are almost parallel to each other, the exchange interactions between the ions work effectively. The directions of ordered moments of Pm and Pr ions, however, subtend an angle of 60(5)°. Hence, the exchange interactions in PrNi probably become complicated. It is plausible that the complicated exchange interactions lessen not only the contact interaction, but also the expectation value $\langle J_z \rangle$ of the $4f$ electrons. This argument stimulates further experiments, such as LTNO of Pm in SmNi.

ACKNOWLEDGMENT

The authors wish to thank Dr. D. Doran for his critical reading of the manuscript.

[1] S. C. Abrahams, J. L. Bernstein, R. C. Scherwood, J. H. Wernick, and H. J. Williams, J. Phys. Chem. Solid **25**, 1067 (1964).
[2] R. E. Walline and W. E. Wallace, J. Chem. Phys. **41**, 1587 (1964).
[3] G. Fillion, D. Gignoux, F. Givord, and R. Lemaire, J. Magn. Magn. Mater. **44**, 173 (1984).
[4] Y. Isikawa, K. Mori, K. Ueno, K. Sato, and K. Maezawa, J. Magn. Magn. Mater. **52**, 434.
[5] K. Nishimura, S. Ohya, K. Heiguchi, S. Muto, Y. Isikawa, and K. Mori, Hyperfine Interact. **78**, 475 (1993).
[6] A. E. Dwight, R. A. Conner, Jr., and J. W. Downey, Acta Crystallogr. **18**, 837 (1965).
[7] K. S. Krane, in *Low-Temperature Nuclear Orientation*, edited by N. J. Stone and H. Postma (North-Holland, Amsterdam, 1986), Chap. 2 and Appendix 5.
[8] N. J. Stone, Hyperfine Interact. **34**, 91 (1987).
[9] *Table of Isotopes*, 7th ed., edited by C. M. Lederer and V. S. Shirley (Wiley, New York, 1978), Appendix 61.
[10] C. J. Ashworth, P. Back, S. Ohya, N. J. Stone, and J. P. White, Hyperfine Interact. **59**, 461 (1990).
[11] N. Shohata, J. Phys. Soc. Jpn. **42**, 1873 (1977).
[12] J. E. Greendan and V. U. S. Rao, J. Solid State Chem. **6**, 387 (1973).
[13] R. J. Elliott and K. W. H. Stevens, Proc. R. Soc. **A218**, 553 (1953).
[14] C. A. Morrison and R. P. Leavitt, in *Handbook on the Physics and Chemistry of Rare Earths*, edited by K. A. Gschneidner, Jr. and L. Eyring (North-Holland, Amsterdam, 1982), Vol. 5, Chap. 46.
[15] B. Bleaney, in *Magnetic Properties of Rare Earth Metals*, edited by R. J. Elliott (Plenum Press, New York, 1972), Chap. 8.
[16] K. Nishimura, I. S. Oliveira, N. J. Stone, Y. Isikawa, K. Shimizu, and K. Sato, Hyperfine Interact. **78**, 153 (1993).
[17] I. S. Oliveira, K. Nishimura, N. J. Stone, Y. Isikawa, D. Zakouchy, and D. Doran, Phys. Rev. B **49**, 11 886 (1994).