# Evidence for nuclear-spin order in double-hcp praseodymium by neutron diffraction

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In search of evidence for nuclear-spin order in double-hexagonal-close-packed Pr metal, neutron-diffraction experiments have been done at millikelvin temperatures. The results of polarization analysis of two magnetic satellites on the [100] reciprocal-lattice axis, and the temperature dependence of the intensities of these satellites have been interpreted in terms of a model which has substantial nuclear-spin order on both hexagonal- and cubic-site atoms.

In singlet electronic-ground-state compounds, nuclear spins have an important role for the onset of long-range magnetic order. Magnetic perturbation by, for example, an exchange field and/or a hyperfine field can induce magnetic moments in the electronic system through the matrix element of the angular momentum between the ground and the excited state, $\alpha \equiv \langle G \mid \mathbf{J} \mid E \rangle$. When the magnitude of the exchange interaction, $2J(\mathbf{Q})$, exceeds a certain threshold, an instability takes place in the ground state and the system undergoes a self-induced spontaneous ordering.[1] Specifically, this threshold is expressed in terms of a parameter $\eta \equiv 4\alpha^2 J(\mathbf{Q})/\Delta$, as $\eta = 1$, where $\Delta$ is the energy separation between $\mid E \rangle$ and $\mid G \rangle$. Taking the hyperfine interaction into account, Murao[2] has shown theoretically that long-range order develops both in the electronic and nuclear-spin systems even when $\eta$ is less than 1. According to his calculation, with $\eta$ close to but slightly less than 1, the magnetization processes of the electrons and the nuclear spins are quite different from each other and from an ordinary Brillouin-type behavior.

Double-hexagonal praseodymium has an $ABAC$ stacking sequence along the $c$ axis. The atoms experience a crystal field of local cubic symmetry in the $A$ layers and of local hexagonal symmetry in the $B$ and $C$ layers. The electronic system, in which the atomic-electronic configuration is $4f^2$ and the ground multiplet is $^3H_4$, has a singlet ground state at both the hexagonal and the cubic sites, and it has been established[3] that the magnitude of the exchange interaction between the hexagonal-site ions is just insufficient by itself for spontaneous ordering. In the first neutron-diffraction experiment on this material at millikelvin temperatures, McEwen and Stirling[4] observed several magnetic satellite reflections originating from a modulated magnetic structure with a fundamental propagation vector of $0.13\mathbf{a}^*$ at temperatures well below 1 K, where $\mathbf{a}^*$ is the reciprocal lattice vector of magnitude $(4\pi/a\sqrt{3})$. An intensity analysis led to the conclusion that the overall magnetic structure is sinusoidal with components parallel and perpendicular to the $a^*$ axis. This magnetic ordering was believed to be driven by only the hexagonal site ions with a marginal exchange interaction in cooperation with the nuclear spins through the hyperfine interaction.[5] Subsequently, Bjerrum Møller *et al.*[6] and Stirling and McEwen[7] measured the temperature dependence of the spectra and the intensities of some of these satellites. These experiments revealed that they exist even at 1 K although the intensity is very weak and the width of the peak is appreciably broader than at lower temperatures. The intensity remains almost constant down to about 100 mK and increases dramatically below about 60 mK, and the peaks become narrow enough at this temperature to represent a true long-range order. Such a behavior is in qualitative agreement with the theoretical prediction by Murao,[2] though the mean-field theory is not concerned with the width of the peak, which relates to the size of spatial fluctuations of the ordering.

It should be noticed that in the neutron experiments on this material which have been made up to the present, the effect of nuclear-spin polarization on the intensity of the observed magnetic peaks has been assumed to be small. No direct evidence by neutron diffraction has been obtained for the existence of the nuclear-spin contribution to the long-range order in this material. On the other hand, in a neutron-diffraction experiment on $PrSn_3$, Kawarazaki *et al.*[8] have demonstrated that pseudomagnetic scattering from polarized nuclear spins of Pr atoms is intense enough to be observed. With this in mind, we have tried in the present work to search for evidence of nuclear-spin order in Pr metal by neutron diffraction.

Interacting with a nucleus with nuclear-spin polarization of $\langle \mathbf{I} \rangle$, a neutron is scattered coherently by the nuclear pseudomagnetic moment[9]

$$\langle \mu_N^* \rangle = \frac{B}{0.27} \langle \mathbf{I} \rangle (\mu_B) , \tag{1}$$

with a scattering amplitude which is expressed in a quantum-mechanical operator form as

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$$p_N = 0.27(\mu_N^* \cdot \sigma) \quad (2)$$

in units of $10^{-12}$ cm, where $B$ is a constant which is characteristic of the nuclear isotope and $\sigma$ is the Pauli operator for the neutron spin. On the other hand, the magnetic scattering length due to an electronic magnetic moment $\mu$ is expressed as

$$p_e = 0.27 f(\kappa)(\mu_\perp \cdot \sigma) , \quad (3)$$

where $\mu_\perp$ is the component of $\mu$ perpendicular to the scattering vector $\kappa$ and $f(\kappa)$ is the form factor of the magnetic electrons. Unlike $p_N$, $p_e$ depends on the scattering vector. Using this fact one can measure, in principle, both $\langle \mu_N^* \rangle$ and $\langle \mu \rangle$ separately. The magnitude of the nuclear pseudomagnetic moment of the $^{141}$Pr nucleus at 0 K has been determined to be[8]

$$\mu_N^*(0) = \frac{BI}{0.27} = -0.17\mu_B . \quad (4)$$

The single crystal for the present experiment is one which was used by McEwen and Stirling[10,11] in earlier studies of uniaxial stress-induced magnetic ordering in praseodymium. The crystal was aligned so that the scattering vector lies in the $a^*$-$c^*$ reciprocal plane, and the diffraction intensities were measured by means of the polarization-analysis technique using the HB1 spectrometer of the High-Flux Isotope Reactor at ORNL. A copper bracket, cut from a single crystal, was soldered to the top of the sample and secured to the cold tip of a dilution refrigerator. Three temperature sensors were attached to a similar bracket soldered to the bottom of the sample. A carbon resistance thermometer was used to measure temperature during the neutron experiment, and it was calibrated against a $^{60}$Co nuclear orientation thermometer and a standard Ge resistance thermometer immediately before mounting the refrigerator on the spectrometer. A small vertical magnetic field of 0.025 T served as a guide for the neutron polarization. The wavelength was 1.529 Å and 40-min collimators were used before and after the sample.

Among several satellite peaks previously observed by McEwen and Stirling,[4] we were interested in the weak peak at the (0.87,0,0) reciprocal position which is one of the satellites of the (100) reciprocal lattice point. An electronic sinusoidal structure with polarization parallel to the $a^*$ axis does not allow a satellite to exist on the $a^*$ axis because $\mu_\perp$ is zero at any position on this axis. In order to explain the existence of the peak at (0.87,0,0), it had been assumed that there is another component of the electronic moment which lies in the crystal $c$ plane and is perpendicular to the $a^*$ axis. However, if one assumes an appreciable amount of nuclear-spin polarization, parallel to the electronic moment, it is not necessary to assume this extra electronic component because the nuclear spin scattering has no restriction due to the geometrical factor.

One can examine whether or not this peak is purely due to nuclear-spin scattering by measuring two peaks along the $a^*$ axis and by examining whether or not the difference in the intensities between them can be explained just in terms of the Lorentz factor without using the electronic form factor. For this purpose, we studied two satellites on both sides of the (100) reciprocal point along the $a^*$ axis.

In Fig. 1, the spectra of the (0.87,0,0) and (1.13,0,0) peaks (hereafter denoted as $-$ and $+$ satellites, respectively) measured at 23 mK are shown with the neutron spin flipper on (yielding spin-flip scattering by the sample) and off (giving non-spin-flip scattering). Before discussing the intensity, we consider the observed flipping of the neutron spin shown in this figure. One can see in the figure that scattering for these two peaks is mostly spin-flip scattering. Indeed, correcting for the imperfect polarization of the incident neutrons, we obtain a value of $1.0 \pm 0.05$ for the ratio of spin-flip scattering to the total scattering. Since the incident neutron polarization was perpendicular to the $a^*$-$c^*$ scattering plane, we definitely conclude that the polarization of the electronic moment and/or the nuclear spins which yields these peaks lies in this plane. Therefore, if this scattering is electronic, then the moment must have a component parallel to the $c^*$ axis. However, if this scattering is due to the nuclear spins, it is possible to explain the observed spin-flip scattering without assuming any extra component of polarization in addition to the component parallel to the $a^*$ axis.

The observed intensities of the two peaks make the situation somewhat complicated. If these two peaks are purely due to nuclear-spin scattering from the hexagonal sites, the intensity ratio, $I^+/I^-$ should be 0.8 taking the Lorentz factor into account, while, if they are due to electronic scattering, the ratio should be 0.7 due to both Lorentz and form factors. However, the observed ratio of the integrated intensities was as small as 0.3. Furthermore, we have found that this intensity ratio is strongly temperature dependent, as shown in Fig. 2.

The observed behavior of the intensities is well reproduced by a model in which not only the hexagonal sites but also the cubic sites participate in the magnetism of this material. In this model, only polarization which is parallel to the $a^*$ axis is required. We consider the following set of sinusoidal waves in the four $c$ planes:

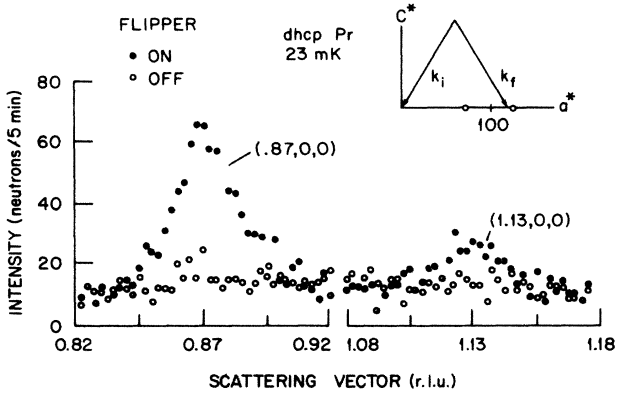


FIG. 1. Diffraction spectra of the (0.87,0,0) and (1.13,0,0) satellites at 23 mK with polarization analysis. The neutron polarization was perpendicular to the $a^*$-$c^*$ reciprocal plane.

$$\begin{aligned}
&\mu^*_{NA}(\mathbf{r})=\mu^*_{NA}\hat{\mathbf{q}}\sin(\mathbf{q}\cdot\mathbf{r}) \text{ for the } A \text{ plane},\\
&\mu^*_{NA'}(\mathbf{r})=\mu^*_{NA'}\hat{\mathbf{q}}\sin(\mathbf{q}\cdot\mathbf{r}) \text{ for the } A' \text{ plane},\\
&\mu^*_{NB}(\mathbf{r})=\mu^*_{NB}\hat{\mathbf{q}}\cos(\mathbf{q}\cdot\mathbf{r}) \text{ for the } B \text{ plane},\\
&\mu^*_{NC}(\mathbf{r})=\mu^*_{NC}\hat{\mathbf{q}}\cos(\mathbf{q}\cdot\mathbf{r}) \text{ for the } C \text{ plane},
\end{aligned} \tag{5}$$

where $\hat{\mathbf{q}}$ is the unit vector in the $a^*$ direction. Equation (5) is a modification of the more general form of the magnetic structure discussed by Bak and Lebech,[12] applied to the (dhcp) double-hcp Nd metal. In this equation only the nuclear-spin polarizations are shown. The electronic magnetic moments with the same structure do not give intensity to these satellite peaks because of the null geometrical factor. The phase difference of $\pi/2$ between the waves on the cubic sites and the hexagonal sites is discussed later.

The structure factors of the $\pm$ satellites for this magnetic structure are calculated to be

$$F^{\pm}(\mathbf{G})=\tfrac{1}{2}[F^h(\mathbf{G})\pm iF^c(\mathbf{G})], \tag{6}$$

where

$$\begin{aligned}
F^h(\mathbf{G})&=0.27(\mu^*_{NB}e^{i\mathbf{G}\cdot\rho_B}+\mu^*_{NC}e^{i\mathbf{G}\cdot\rho_C}),\\
F^c(\mathbf{G})&=0.27(\mu^*_{NA}e^{i\mathbf{G}\cdot\rho_A}+\mu^*_{NA'}e^{i\mathbf{G}\cdot\rho_{A'}}),
\end{aligned} \tag{7}$$

$\mathbf{G}$ and $(\rho_A\rho_{A'}\rho_B\rho_C)$ being the reciprocal-lattice position associated with the satellites and the atomic positions in the unit cell, respectively. We now make the following assumptions: (i) In the planes which have the same local symmetry, the amplitudes are the same:

$$|\mu^*_{NA}|=|\mu^*_{NA'}| \text{ and } |\mu^*_{NB}|=|\mu^*_{NC}|;$$

(ii) $\mu^*_{NB}=-\mu^*_{NC}$ $(\equiv\mu^{*h}_N)$ have nonzero $F^h$ for $\mathbf{G}=(2\pi/c)(001)$; (iii) $\mu^*_{NA}=\mu^*_{NA'}$ $(\equiv\mu^{*c}_N)$ have nonzero $F^c$ for $\mathbf{G}=(4\pi/a\sqrt{3})(100)$. Using $\rho_A=(000)$, $\rho_B=(\frac{2}{3}\frac{1}{3}\frac{1}{4})$, $\rho_{A'}=(00\frac{1}{2})$, and $\rho_C=(\frac{1}{3}\frac{2}{3}\frac{3}{4})$, we obtain

$$F^{\pm}(100)=-0.27i[(\sqrt{3}/2)\mu^{*h}_N\mp\mu^{*c}_N]. \tag{8}$$

The ratio of the satellite intensities to the (100) nuclear intensity is given by

$$\begin{aligned}
I^{\pm}/I^{100}&=\frac{\frac{1}{3}L^{\pm}|F^{\pm}|^2}{L^{100}|F^{100}|^2}\\
&=\frac{0.27^2L^{\pm}[(\sqrt{3}/2)\mu^{*h}_N\mp\mu^{*c}_N]^2}{3L^{100}b^2},
\end{aligned} \tag{9}$$

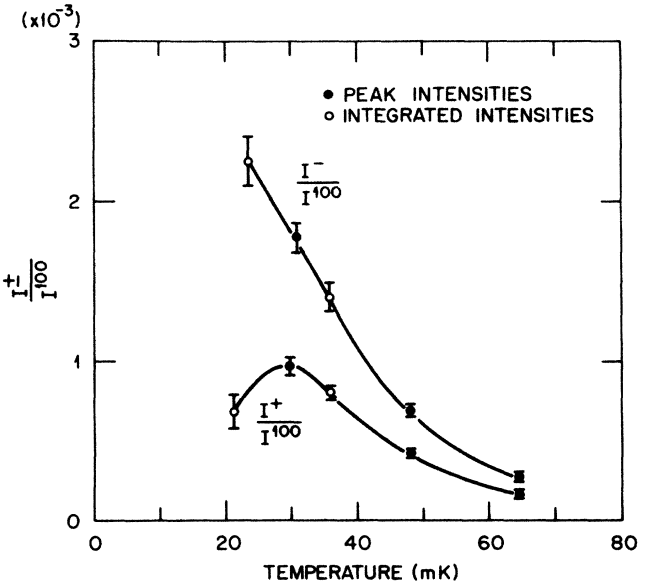


FIG. 2. Temperature dependences of the (0.87,0,0) and (1.13,0,0) intensities normalized to the (100) intensity. The 23-mK data were obtained in the polarization-analysis mode; the other points were obtained with an unpolarized beam. The peak-intensity ratios were calculated as $[I^{\pm}_p(T)/I^{\pm}_p(36)][I^{\pm}(36)/I^{100}(36)]$ where $I_p$ is a peak intensity and $I$ is an integrated intensity.

where $b$ is the coherent scattering length of Pr with no nuclear-spin polarization, $L$ is the appropriate Lorentz factor, and the factor of $\frac{1}{3}$ is due to magnetic domains assumed to be equally distributed. Using the data of Fig. 2 and Eq. (9), the pseudomagnetic moments were obtained as a function of temperature. These results are given in Table I. It should be remembered that these numbers represent the amplitudes of modulations for a set of sites rather than the pseudomagnetic moment on a particular site. The corresponding nuclear polarizations were obtained by dividing the moments of Table I by $|\mu^*_N(0)|=0.17\mu_B$.[8]

While we have not observed the electronic polarization in this experiment, it is certainly present. We expect the nuclear and electronic polarizations at each atomic site to be coupled via the hyperfine interaction with the relationship given by

$$P=B_{5/2}\left[\frac{A\langle J_z\rangle I}{kT}\right], \tag{10}$$

TABLE I. Temperature dependence of the amplitudes of the modulated nuclear pseudomagnetic moments on hexagonal and cubic sites.

| $T$ (mK) | $\lvert\mu^{*h}_N\rvert(\mu_B)$ | $\lvert\mu^{*c}_N\rvert(\mu_B)$ | $P^h$ | $P^c$ |
|---|---|---|---|---|
| 23.7 | 0.124±0.004 | 0.020±0.004 | 0.729±0.023 | 0.118±0.024 |
| 30.7 | 0.120±0.002 | 0.0095±0.0021 | 0.706±0.011 | 0.056±0.012 |
| 35.6 | 0.107±0.002 | 0.0070±0.0018 | 0.629±0.011 | 0.041±0.010 |
| 48.0 | 0.076±0.002 | 0.0040±0.0014 | 0.447±0.011 | 0.024±0.008 |
| 64.5 | 0.048±0.001 | 0.0020±0.0011 | 0.282±0.006 | 0.012±0.006 |

where $B_I$ is the Brillouin function and $A$ is the hyperfine interaction coefficient. Taking $A=1093$ MHz (Ref. 8) and $\mu=0.8\langle J_z\rangle$, we obtain

$$P(T)=B_{5/2}\left[\frac{164\mu(T)}{T}\right], \tag{11}$$

with $\mu$ in Bohr magnetons and $T$ in mK. Using the results in Table I and Eq. (11), the amplitudes of the electronic modulations shown in Table II were obtained. The value for $\mu^h$ at the lowest temperature seems inconsistent with the other numbers. The 23.7-mK data were obtained with the polarization-analysis experimental arrangement and the remainder of the data were obtained with an unpolarized beam. There may have been a systematic error in one or the other of these arrangements resulting in this inconsistency.

From measurements on the ($q$03) satellite, Bjerrum Møller *et al.*[6] obtained a value of $0.36\mu_B$ at 40 mK for the amplitude of the hexagonal electronic moment. This is in remarkable agreement with the corresponding value from Table II; however, their result was obtained without considering the nuclear polarization. They assumed the satellite intensity to be given by

$$I_{q03}\sim m^2f^2\cos^2\alpha\ , \tag{12}$$

where $m$ is the hexagonal moment, $f$ is the $4f$ form factor, and $\alpha$ is the angle between the $c$ axis and the scattering vector. In our model, this relationship is replaced by

$$I_{q03}\sim(\mu^h)^2f^2\cos^2\alpha+(\mu_N^{*h})^2-2\mu^h\mu_N^{*h}f\cos^2\alpha\ , \tag{13}$$

where the negative sign on the third term is determined by the sign of $\mu_N^*$. Using our results for $\mu^h$ and $\mu_N^{*h}$, we calculate a value for $m$ in their experiment of $0.25\mu_B$ at 40 mK. Alternatively, if we adopt their value of $0.36\mu_B$ for $m$ and our value of 0.096 for $\mu_N^{*h}$, we deduce that $\mu^h=0.46\mu_B$. This should be compared with $\mu^h=0.35\mu_B$ obtained by interpolation in Table II. The difference is probably due to temperature uncertainties in the two experiments. As a suggestion for further work, our results and Eq. (13) indicate that the ($q$03) intensity should go through a maximum at about 35 mK.

It is interesting to note that if Eq. (10) is valid, either the nuclear or electronic modulation must deviate from a pure sinusoid when $P$ is large enough for the relationship to be nonlinear. The dividing line between linear and nonlinear behavior occurs at about $B_{5/2}=0.2$, so that the hexagonal-site modulation is definitely in the nonlinear regime. For a sinusoidal modulation of the electronic moments, the nuclear polarization will show a distorted wave which evolves toward a square wave in the limit of full nuclear polarization. We have calculated the influence of such a distortion on our intensities and conclude that at our lowest temperature the actual maximum nuclear polarization on the hexagonal sites would be 6% lower than given in Table I. A corollary is that a peak at $(1\pm3q,0,0)$ should appear, but this would be very weak.

TABLE II. Temperature dependence of the amplitudes of the modulated electronic moments deduced from Eq. (11).

| $T$ (mK) | $\mu^h(\mu_B)$ | $\mu^c(\mu_B)$ | $\mu^c/\mu^h$ |
|---|---|---|---|
| 23.7 | 0.32±0.02 | 0.036±0.007 | 0.113±0.023 |
| 30.7 | 0.38±0.01 | 0.022±0.005 | 0.057±0.013 |
| 35.6 | 0.37±0.01 | 0.019±0.005 | 0.051±0.013 |
| 48.0 | 0.31±0.01 | 0.015±0.005 | 0.048±0.014 |
| 64.5 | 0.25±0.01 | 0.010±0.005 | 0.040±0.020 |

Our satellite peaks were definitely broader than the nuclear (100) peak. In units of $G_{100}$, the $(100)^-$ peak had a width of 0.027 at 23.7 and 35.6 mK while the (100) peak had a width of 0.018. This sort of behavior was observed by Bjerrum Møller *et al.*,[6] but the effect was much smaller and occurred at higher temperature. The implication is that our nuclear ordering was not truly long range.

When one discusses the possibility of having a magnetic moment on the cubic-site atoms, the neutron diffraction experiment by Lebech and Rainford[13] is very suggestive. They have measured at 4.2 K the electronic moments at both cubic and hexagonal sites which are induced by an external field in both parallel and perpendicular directions to the $c$ axis and found that the susceptibility perpendicular to the $c$ axis at the cubic sites is one half of that at the hexagonal sites. We therefore regard it as quite reasonable that the cubic-site atoms yield an induced electronic moment together with nuclear-spin polarization in the exchange field from the ordered hexagonal-site moments.

If one aims to explain the observed intensities using a similar model but with purely electronic moments, one must assume a component of electronic moment which is parallel to the $c$ axis at both the hexagonal and the cubic sites. However, it is immediately obvious from the crystal-field energy level scheme[3] that the susceptibility of the hexagonal-site ions for fields along the $c$ axis is extremely small at low temperatures. Indeed, this has been confirmed in a number of experiments.[13,14]

It is also very difficult to produce the observed intensities from a model which includes electronic and nuclear spins located only on the hexagonal sites, without making the nonphysical assumption that the electronic moment has its largest component in the $a^*$ direction while the nuclear moment has its largest component in the $c^*$ direction. Thus we conclude that the satellites along the $a^*$ axis are due to sinusoidal modulations of the nuclear spins on both the hexagonal and cubic sites, with polarization along the $a^*$ axis.

In low-temperature specific-heat experiments on a single crystal of Pr, Lindelof, Miller, and Pickett[15] and Eriksen, Forgan, Murihead, and Young[16] have observed a steep increase in the specific heat with decreasing temperature around 30 mK and attributed this heat-capacity anomaly to a long-range ordering of the nuclear spins. In the present experiment we found that the intensity of the $(100)^{\pm}$ satellites was too small to measure at 200 mK, while in the experiments of McEwen and Stirling,[4,5] the $(001)^{\pm}$ satellites, which include both electronic and nuclear-spin contributions, have nonzero intensity even at 1 K. This fact indicates that appreciable nuclear polarization starts at much lower temperatures than the electronic polarization. The $(100)^{\pm}$ satellites appear around 100 mK and grow rapidly at near 40 mK.

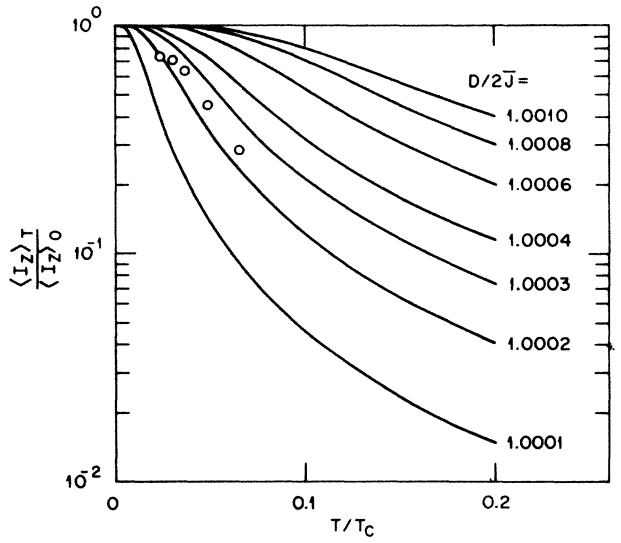


FIG. 3. Comparison of the observed nuclear polarization on the hexagonal sites with calculations by Murao (see Ref. 2 for description of method). $D$ is the energy separation between a singlet ground state and the first excited levels, and $\bar{J}$ is the exchange energy.

McEwen *et al.*[5,10,11] have observed a large enhancement of the entire magnetism of Pr upon applying an external uniaxial stress on the crystal along the $a$ axis. With a stress of 98 MPa, they observed the $(100)^{\pm}$ satellites to exist at temperatures as high as 7 K, where one cannot expect any appreciable nuclear polarization. Therefore, these $(100)^{\pm}$ satellites have been ascribed to a stress-induced electronic moment perpendicular to the $a^*$ axis on the hexagonal sites, presumably induced by the stress. This can be tested by studying these satellites under stress using polarization analysis.

The particular model we have used in analyzing our data is consistent with the idea that the cubic-site polarization is driven by the exchange field set up by the modified electronic moments on the hexagonal sites.[17] The exchange field at an $A$ atom is written as

$$\mathbf{H}_A = \sum_i J(\mathbf{r}_i)\mu^h(\mathbf{r}_A + \mathbf{r}_i) , \tag{14}$$

where $\mathbf{r}_i$ is the set of vectors connecting site $A$ to its nearest hexagonal neighbors. Considering the symmetry around the $A$ atom, we assume that $J(\mathbf{r}_i)=J$ for all $\mathbf{r}_i$. Using Eq. (5) we can rewrite Eq. (14) as

$$\mathbf{H}_A = -4\mu^h J\hat{\mathbf{q}}\sin(\mathbf{q}\cdot\mathbf{r}_A)\sin(2\pi\beta/3)[1-\cos(2\pi\beta/3)] , \tag{15}$$

where $\mathbf{q}=\beta\mathbf{a}^*$. This equation accounts for the phase difference of $\pi/2$ between the waves in the $A$ and $B$ or $C$ planes. Because the same equation is obtained for the exchange field at the $A'$ atom, the waves on the $A$ and the $A'$ planes should be in phase. This was assumed in Eq. (5). It also predicts there should be no induced moment on the $A$ atom if the magnetic structure were such that all the hexagonal-site atoms have the same magnitude of moment, for example, if $\beta=\frac{3}{2}$. If Eq. (15) is valid, then we expect the cubic electronic moment to be proportional to the hexagonal electronic moment at all temperatures. The final column of Table II shows this to be true within experimental error except for the lowest temperature.

Murao[2] has shown that the temperature dependence of the nuclear and electronic polarizations for systems similar to praseodymium can be very sensitive to the ratio of crystal field and exchange energies. While his numerical calculations ($J=1$, $I=\frac{1}{2}$, and ferromagnetic exchange) are not directly applicable to praseodymium, his theory contains the essential physics of the problem and his curves of electronic polarization versus reduced temperature for particular parametric values are very similar to the unusual behavior shown by praseodymium. In Fig. 3 we show our measured nuclear polarization for the hexagonal sites together with a family of curves calculated by Murao for the nuclear polarization. In plotting the experimental points, we have used $T_c=1$ K, but it should be noted that there is a large uncertainty in this number. Murao had already concluded that the case of $D/2\bar{J}=1.0002$ is descriptive of praseodymium, and the present results confirm this conclusion.

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[1]G. T. Trammell, Phys. Rev. **131**, 932 (1963).

[2]T. Murao, J. Phys. Soc. Jpn. **50**, 3240 (1981).

[3]J. G. Houmann, B. D. Rainford, J. Jensen, and A. R. Mackintosh, Phys. Rev. B **20**, 1105 (1979).

[4]K. A. McEwen and W. G. Stirling, J. Phys. C **14**, 157 (1981).

[5]K. A. McEwen, W. G. Stirling, and C. Vettier, in *Crystalline Electric Field Effects in f-Electron Magnetism*, edited by R. P. Guertin, W. Suski, and Z. Zolnierek (Plenum, New York, 1982), p. 57.

[6]H. Bjerrum Møller, J. Z. Jensen, M. Wulff, A. R. Mackintosh, O. D. McMasters, and K. A. Gschneidner, Jr., Phys. Rev. Lett. **49**, 482 (1982).

[7]W. G. Stirling and K. A. McEwen, in *Multicritical Phenomena*, edited by R. Pynn and A. T. Skejltorp (Plenum, New York, 1984), p. 213.

[8]S. Kawarazaki, N. Kunitomi, Y. Morii, H. Suzuki, R. M. Moon, and R. M. Nicklow, Solid State Commun. **49**, 1147 (1984).

[9]A. Abragam, G. L. Bacchella, H. Glattli, R. Meriel, J. Piesvaux, and M. Pinot, C. R. Acad. Sci. Ser. B **274**, 423 (1972).

[10]K. A. McEwen, W. G. Stirling, and C. Vettier, Phys. Rev. Lett. **41**, 343 (1978); Physica (Utrecht) **120B**, 152 (1983); J. Magn. Magn. Mater. **31-34**, 599 (1983).

[11]J. Jensen, K. A. McEwen, and W. G. Stirling, Phys. Rev. B **35**, 3327 (1987).

[12]P. Bak and B. Lebech, Phys. Rev. Lett. **40**, 800 (1978).

[13]B. Lebech and B. D. Rainford, J. Phys. (Paris) Colloq. **32**, C1-370 (1971).

[14]K. A. McEwen, G. J. Cock, L. W. Roeland, and A. R. Mackintosh, Phys. Rev. Lett. **30**, 287 (1973).

[15]P. E. Lindelof, I. E. Miller, and G. R. Pickett, Phys. Rev. Lett. **35**, 1297 (1975).

[16]M. Eriksen, F. M. Forgan, C. M. Murihead, and R. C. Young, J. Phys. F **13**, 929 (1983)

[17]R. M. Moon, J. W. Cable, and W. C. Koehler, J. Appl. Phys. **35**, 1041 (1964).