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TWO-DIMENSIONALLY-MODULATED, MAGNETIC

STRUCTURE OF NEODYMIUM METAL

by

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**Abstract**

The magnetic structure of dhcp Nd was deter-
mined by combining the results of neutron dif-
fraction studies with the results of Landau sym-
metry arguments and renormalization group theory.
The incipient magnetic order is a two-dimen-
sional, incommensurably modulated structure
("triple-$\vec{q}$" structure). The ordering is accom-
panied by a lattice distortion that forms a
similar pattern. The modulation vector decreases
from slightly above $1/7\ \hat{b}_1$ at $T_N$ to $1/8\ \hat{b}_1$ at
$\sim 10$ K. The moments at the hexagonal sites lie
in the basal plane while those at the cubic
sites lie predominantly along the hexagonal axis.
The size of the moments on the cubic sites is
approximately 25% of that of the moments on the
hexagonal sites, with a ratio of three between
the moment components along the hexagonal axis
and those in the basal plane.

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The crystal structure of Nd is hexagonal with a stacking
sequence ABA'C along the c-axis ($\hat{z}$). Moon et al.[1] reported
the first neutron scattering data on Nd. They found satel-
lites in the elastic spectrum displaced from reciprocal lat-
tice points ($\vec{\tau}$) by vectors $\pm\vec{q}_k$, k = 1,2,3 in the three equi-
valent $\hat{b}_1$ directions. They proposed a model for the incipient
magnetic ordering that persists down to ~ 7.5 K[1-3]. In this
model only the spins in the B and C layers order, and the
model has several inadequacies when compared to the experimen-
tal results. Most important, it predicts zero intensities of
satellites on the $\hat{b}_1$-axes and identical intensities of satel-
lites at $\vec{\tau}+\vec{q}_k$ and $\vec{\tau}-\vec{q}_k$. Small, but finite intensities were
observed on the $\hat{b}_1$-axes, and, likewise, different intensities
were observed for satellites at $\vec{\tau}\pm\vec{q}_k$. Our neutron diffraction
measurements, performed at 10 K on single crystals of Nd using
the steady-state reactor DR3 at Risø, agree with the results
described by Moon et al. In addition, we studied the tempera-
ture dependence of selected satellites near $T_N$.

From the temperature dependences of {q,0,$\ell$} satellite in-
tensities, we conclude that the transition at $T_N$ is of second
order (Fig. 1). According to one of the Landau rules, the sym-
metry-breaking order parameter should then transform as an
irreducible representation of the space group of the paramag-
netic phase. The star of $\vec{q}_1$ consists of the six equivalent
basal plane vectors $\pm\vec{q}_k$ where $q_k$ varies from ~ 0.144 at $T_N$ =
19.9 K to $q_k$ ≈ 1/8 at T ~ 7.5 K. The point group that leaves
$\vec{q}_1$ invariant is $C_{2v}$. This group has four one-dimensional re-
presentations, so the order parameter has n = 6×1 components

denoted $\psi(\pm\vec{q}_k) = M_k\exp(\pm i\alpha_k)$, where $k = 1,2,3$. The most general structure described by the order parameters $M_k$ and $\alpha_k$ is linear combinations of the terms

$$\vec{\mu}_A^k(\vec{r}) = (\mu_c\hat{q}_k - \mu_z\hat{z}) \cos(\vec{q}_k\vec{r} + \alpha_k)$$

$$\vec{\mu}_{A'}^k(\vec{r}) = (\mu_c\hat{q}_k + \mu_z\hat{z}) \cos(\vec{q}_k\vec{r} + \alpha_k)$$

$$\vec{\mu}_B^k(\vec{r}) = \mu_h\hat{q}_k \cos(\vec{q}_k\vec{r} + \theta/2 + \alpha_k)$$

$$\vec{\mu}_C^k(\vec{r}) = \mu_h\hat{q}_k \cos(\vec{q}_k\vec{r} - \theta/2 + \alpha_k) ,$$

(1)

where the parameters $\mu_h$, $\mu_c$, $\mu_z$ and $\theta$ must be determined experimentally from a neutron diffraction study. In the Landau theory, the free energy is expanded in terms of the components of the order parameter:

$$F = r \sum_k M_k^2 + u \sum_k M_k^4 + v \sum_{k>j} M_k^2 M_j^2 .$$

(2)

By minimizing F, we find[2] that the "phase diagram" consists of three different regions, one region of first-order transition (III) and two regions (I and II) of second-order transition with different ordered states. In region I, the ordered state has either $M_1$, $M_2$ or $M_3 \neq 0$ ("single-$\vec{q}$" structure). By choosing $\mu_c = \mu_z = 0$ and $\theta = 180^{\circ}$, we obtain the structure of Moon et al. In region II the ordered state has $M_1 = M_2 = M_3 \neq 0$, i.e., the three different equivalent q-vectors are present simultaneously ("triple-$\vec{q}$" structure). In the critical region, Landau theory is insufficient, and near the phase transition the coefficients u and v should be replaced by the renormalized values obtained by repeated applications of the renormalization group transformation. At a second-order transition, u and v should converge to a stable fixed point. Since the only stable fixed point is in region II, and we have found a second-order transition experimentally, we conclude[2] that the

ordered state must have the "triple-$\vec{q}$" structure.
The most general "triple-$\vec{q}$" structure is a superposition
of the "single-$\vec{q}$" components in (1), i.e. $\vec{u}_J(\vec{r}) = \sum_k \vec{u}_J^k(\vec{r})$,
$J = A,B,A',C$; which only depend on $\alpha_k$ through $\alpha = \sum_k \alpha_k$. The
sixth-order term in $F$ fixes $\alpha$ to either $0^O$ or $90^O$.

The magnetic neutron diffraction cross sections of a
pair of satellites at $(\vec{\tau} \pm \vec{q}_k)$ are identical for the "single-$\vec{q}$"
and the "triple-$\vec{q}$" structures. To distinguish experimentally
between the "single-$\vec{q}$" and the "triple-$\vec{q}$" structures, it is
necessary to consider the coupling to the lattice[2]. The spin-
lattice coupling introduces two additional terms in the free
energy expansion; firstly, the usual magnetostriction with
wave vector $2\vec{q}_k$, and secondly lattice distortions that are
$90^O$ out of phase with the magnetization and with periodici-
ties given by $\vec{q}_k$. The latter term involves coupling to two
different components $\psi(\vec{q}_1)$ and $\psi(\vec{q}_2)$ of the order parameter,
thus it is active only in the "triple-$\vec{q}$" state. To visualize
the resulting structure, we have plotted the basal plane B-
site spins on top of the distorted B-site lattice (Fig. 2).

The distortion gives rise to satellites in the diffrac-
tion spectrum which generally coincide with the magnetic sat-
ellites. However, while the magnetic satellites $(h\pm q,0,0)$
vanish, the corresponding structural satellites are allowed.
The existence of $(h\pm q,0,0)$ satellites is thus direct evidence
of the "triple-$\vec{q}$" structure. As the lattice distortion $(u(\vec{q}_k))$
couples to second order in the primary order parameter, its
temperature dependence in the critical region should be
$|u(\vec{q}_k)| \propto (T_N-T)^{2\beta}$, in contrast to the primary order para-

meter $|\psi(\vec{q}_k)| \propto (T_N-T)^\beta$.

We compared the measured temperature dependences of the $(1-q,0,0)$ and the $(q,0,3)$ satellites (Fig. 1A). The Néel temperature was estimated to be $(19.9\pm0.1)$ K both from the peak in the elastic scattering away from the $(q,0,3)$ Bragg point and from the divergence in the width of the satellites close to $T_N$ (Fig. 1B). The critical scattering, which was subtracted from the $(q,0,3)$ intensity below $T_N$, was estimated from the $(q,0,3)$ intensity above $T_N$ using the scaling laws. After a few iterations, $\beta$ converged at $\beta = (0.36\pm0.02)$, in agreement with the theoretical value of $\beta \sim 0.38$ calculated to second order in $\epsilon$. The $(1-q,0,0)$ intensity is consistent with $\beta' \sim 1\pm0.3 \sim 2\beta$, thus confirming that the peak arises from a second-order coupling to the order parameter that <u>only exists</u> for the "triple-$\vec{q}$" <u>structure</u>. The possibility of multiple scattering effects involving two magnetic satellites was ruled out by experimental tests and calculation.

The four parameters giving the magnitude and the phase of the spins in the different layers, and the four parameters giving the corresponding lattice distortions were determined from the intensities at 10 K of $\sim 40$ independent satellites. For the common phase $\alpha = 0°$, we found the maximum moments of the "triple-$\vec{q}$" structure to be $\mu_{Mh} \sim 2.36 \ \mu_B$, $\mu_{Mc} \sim 0.4 \ \mu_B$, $\mu_{Mz} \sim 1.08 \ \mu_B$, and $\theta \sim 180°$. For $\alpha = 90°$, we found $\mu_{Mh} \sim 2.54 \ \mu_B$, $\mu_{Mc} \sim 0.3 \ \mu_B$, $\mu_{Mz} \sim 0.72 \ \mu_B$, and $\theta \sim 180°$. The associated lattice distortions are of the order of a few per cent. The data suggest that the distortions are confined to the basal planes and restricted to distortions of either the hexagonal

or of the cubic sites.

## References

1) MOON, R.M., CABLE, J.W., and KOEHLER, W.C., J. Appl. Phys. 33 (1964) 1041.

2) BAK, P. and LEBECH, B., Phys. Rev. Lett. 40 (1978) 800 and references therein.

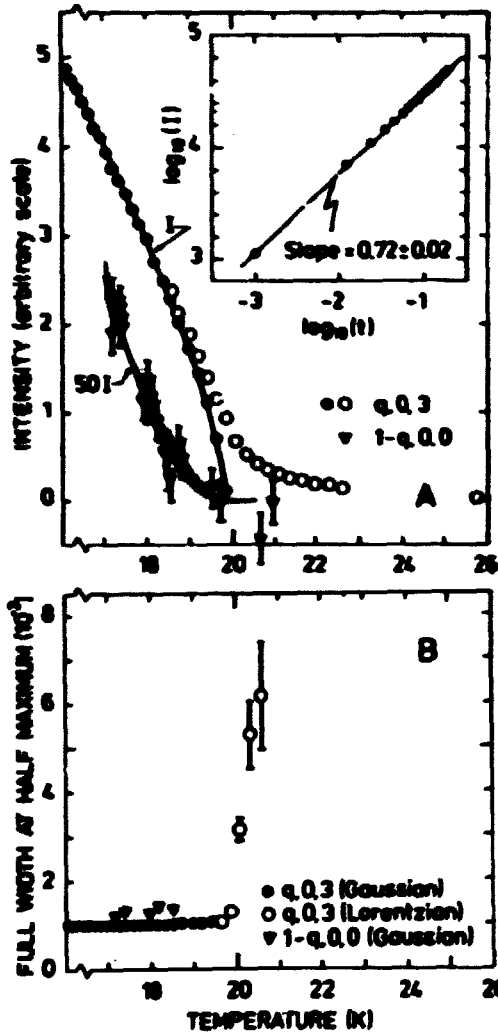3) LEINKKERI, J. and PALMER, S.B., J. Phys. F: Metal Phys. 7 (1977) 15.

Fig. 1. A) Temperature dependences of the neutron intensities of the structural and the magnetic satellites. The solid lines are the results of least squares fits to power laws. The hatched area corresponds to a 25% variation of $\beta'$ describing the temperature dependence of (1-q,0,0). The open circles show the (q,0,3) intensities before subtraction of the critical scattering that persists up to ~ 27 K in agreement with Lebkkeri and Palmer[3]. B) The full width at half maximum of the satellites. The increased width of (q,0,3) above ~ 19.7 K is caused by critical scattering. The increased width of (1-q,0,0) is a resolution effect.
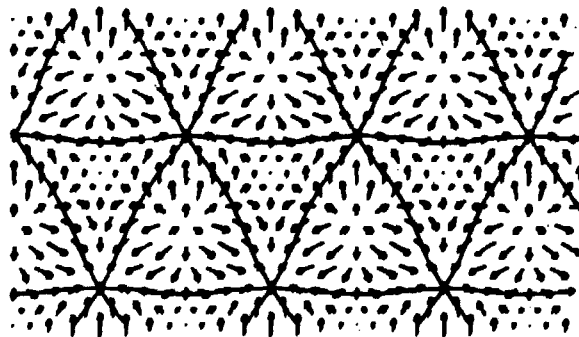


Fig. 2. The magnetic structure of neodymium for $\alpha = 90°$. The basal plane components of the spins in the B layers are centered at the atomic positions of the distorted lattice (thin lines). For illustrative purposes, the amplitudes of the lattice distortion are 0.2 $\delta_x$.