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Publication date:
1978

Document Version
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Citation (APA):
Lebech, B., & Bak, P. (1978). *Two-dimensional-modulated, magnetic structure of neodymium metal*. Risø National Laboratory. Risø-M No. 2124

Risø

Risø-M-2124

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Title and author(s)

TWO-DIMENSIONALLY-MODULATED, MAGNETIC STRUCTURE OF NEODYMIUM METAL

by

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Date 29 July 1978

Department or group

Physics Department

Group's own registration number(s)

6 pages + 0 tables + 2 illustrations

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Abstract

The magnetic structure of dhcp Nd was determined by combining the results of neutron diffraction studies with the results of Landau symmetry arguments and renormalization group theory. The incipient magnetic order is a two-dimensional, incommensurably modulated structure ("triple-$\vec{q}$" structure). The ordering is accompanied 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 satellites in the elastic spectrum displaced from reciprocal lattice points ($\vec{\tau}$) by vectors $\pm\vec{q}_k$, k = 1,2,3 in the three equivalent $\hat{b}_1$ directions. They proposed a model for the incipient magnetic ordering that persists down to $\sim$ 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 experimental results. Most important, it predicts zero intensities of satellites on the $\hat{b}_1$-axes and identical intensities of satellites 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 temperature dependence of selected satellites near T_N.

From the temperature dependences of {q,0,ℓ} satellite intensities, we conclude that the transition at T_N is of second order (Fig. 1). According to one of the Landau rules, the symmetry-breaking order parameter should then transform as an irreducible representation of the space group of the paramagnetic phase. The star of $\vec{q}_1$ consists of the six equivalent basal plane vectors $\pm\vec{q}_k$ where q_k varies from $\sim$ 0.144 at T_N = 19.9 K to $q_k \simeq 1/8$ at T $\sim$ 7.5 K. The point group that leaves $\vec{q}_1$ invariant is C_{2v}. This group has four one-dimensional representations, 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 α_k is linear combinations of the terms

$$\begin{aligned}
\vec{\mu}^k_A(\vec{r}) &= (\mu_c \hat{q}_k - \mu_z \hat{z}) \cos(\vec{q}_k \vec{r} + \alpha_k) \\
\vec{\mu}^k_{A'}(\vec{r}) &= (\mu_c \hat{q}_k + \mu_z \hat{z}) \cos(\vec{q}_k \vec{r} + \alpha_k) \\
\vec{\mu}^k_B(\vec{r}) &= \mu_h \hat{q}_k \cos(\vec{q}_k \vec{r} + \theta/2 + \alpha_k) \\
\vec{\mu}^k_C(\vec{r}) &= \mu_h \hat{q}_k \cos(\vec{q}_k \vec{r} - \theta/2 + \alpha_k) \ ,
\end{aligned} \qquad (1)$$

where the parameters μ_h, μ_c, μ_z and θ 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:

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

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 <u>either</u> 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. $\vec{\mu}_J(\vec{r}) = \sum_k \vec{\mu}_J^k(\vec{r})$, $J = A, B, A', C$; which only depend on α_k through $\alpha = \sum_k \alpha_k$. The sixth-order term in F fixes α to either 0° or 90°.

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° out of phase with the magnetization and with periodicities 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 diffraction spectrum which generally coincide with the magnetic satellites. 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 parameter $|\psi(\vec{q}_k)| \propto (T_N-T)^{\beta}$.

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±0.1) 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, β converged at β = (0.36±0.02), in agreement with the theoretical value of $\beta \sim 0.38$ calculated to second order in ϵ. 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 only exists for the "triple-$\vec{q}$" structure. The possibility of multiple scattering effects involving two magnetic satellites was rules 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 $\alpha = 0^{\circ}$, 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 $\theta \sim 180^{\circ}$. For $\alpha = 90^{\circ}$, we found $\mu_{Mh} \sim 2.54\ \mu_B$, $\mu_{Mc} \sim 0.3\ \mu_B$, $\mu_{Mz} \sim 0.72\ \mu_B$, and $\theta \sim 180^{\circ}$. 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

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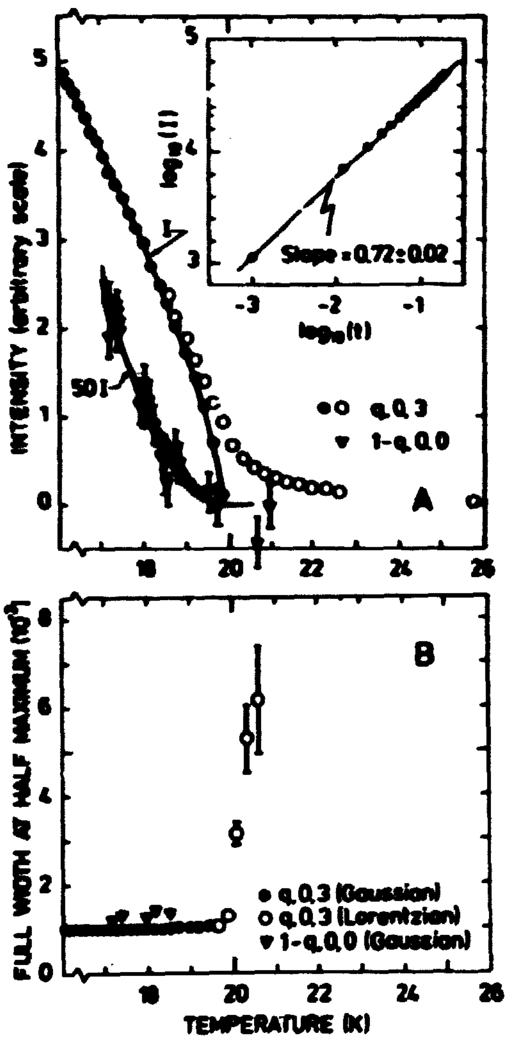

Fig. 1. A) Temperature dependences of the neutron intensities of the structural and the magnetic satellites. The solid lines are the results of least squares fits to power laws. The hatched area corresponds to a 25% variation of β' describing the temperature dependence of (1-q,0,0). The open circles show the (q,0,3) intensities before subtraction of the critical scattering that persists up to ∼ 27 K in agreement with Lenkkeri 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.

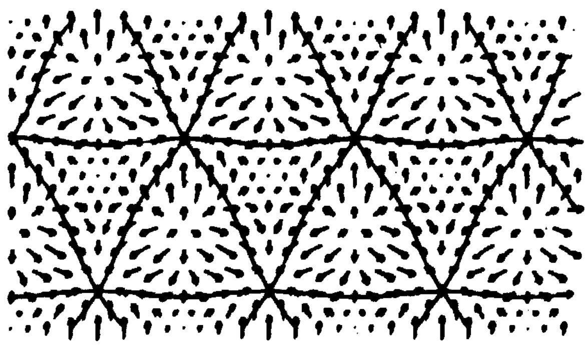

Fig. 2. The magnetic structure of neodymium for α = 90°. The basal plane components of the spins in the B layers are centered at the atomic positions of the distorted lattice (thin lines). For illustrative purposes, the amplitudes of the lattice distortion are 0.2 $\hat{b}_k$.