X-Ray and Neutron Scattering Study of the Magnetic Structure of Neodymium Metal

B. Lebech and J. Als-Nielsen
Physics Department, Risø National Laboratory, DK-4000 Roskilde, Denmark

and

K. A. McEwen
Physics Department, University of Salford, Salford M5 4WT, United Kingdom
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A combined x-ray and neutron diffraction study has shown that the so-called "triple-$\vec{q}$" structure is not the correct model of the magnetic structure of neodymium. The x-ray data showed only the Bragg reflections originating from the double-hcp lattice. Hence, all additional reflections observed below T_N by neutrons are of magnetic origin. Additional neutron-diffraction data have shown that the magnetic structure must be described by modulation wave vectors with components both parallel and perpendicular to the ⟨100⟩ axes.

The magnetic structure of the rare-earth element neodymium (double-hcp structure) is rather complex and has remained an unsolved puzzle for more than a decade. Neutron diffraction studies below 20 K of different single crystals of neodymium performed by Moon, Cable, and Koehler,[1] Shapiro and Sinha,[2] and Lebech and Rainford[3] have all shown Bragg reflections of presumed magnetic origin. Some of these reflections, however, cannot be explained by the originally proposed model[1] of the magnetic structure, a longitudinal modulation of the magnetic moments in the basal planes ("single-$\vec{q}$" structure). Furthermore, it is known from experiment that the transition from the paramagnetic to the magnetically ordered state is continuous. This observation is in contrast to the modern theory of phase transitions (renormalization-group theory) according to which the transition should be discontinuous (first order) if the single-$\vec{q}$ structure is correct. Recently, it was shown by Bak and Lebech[4] that the discrepancy between theory and experiment could be removed by assuming that the structure just below the transition is a superposition of three coexisting modulated magnetic structures ("triple-$\vec{q}$" structure). With this assumption, the transition predicted by theory becomes continuous as observed. In addition, by assuming that the triple-$\vec{q}$ structure couples to the crystal lattice, the previously unexplained Bragg reflections and their temperature dependence could be interpreted as being due to lattice distortions. A definitive test of this hypothesis may be done by comparing neutron diffraction data—containing information about both the magnetic structure and the postulated lattice distortion—to x-ray diffraction data that contains only information about the crystal lattice.

The present note reports the results of an x-ray diffraction study of neodymium in a search for Bragg reflections originating from lattice distortions. The results were negative and this led to an extended collection of neutron diffraction data from neodymium with the result that previously unobserved Bragg reflections of magnetic origin were observed. Some of these results are also reported in this note but a new model of the magnetic structure of neodymium has not yet materialized.

The sample used for the experiments was a single-crystal plate (4 mm×4 mm×0.5 mm) which had been polished and etched to obtain a shiny surface for the x-ray experiment. The crystal was mounted with either the [1$\bar{2}$0] or the [001] axis perpendicular to the scattering plane in a cryostat with beryllium windows transparent in x-ray and neutron radiation.

Figure 1 shows a comparison of neutron and x-ray diffraction data obtained below the magnetic ordering temperature $T_N = 19.9$ K. The neutron diffraction data [Fig. 1(a)] was obtained using 13.6 meV neutrons and a cold neutron spectrometer with a large signal-to-background ratio. The x-ray experiments [Fig. 1(b)] were carried out with use of an x-ray spectrometer and molybdenum $K\alpha$ x rays generated by a 12-kW Rigaku rotating anode generator. The exit beam was monochromatized by Bragg scattering from a pyrolytic graphite crystal using the (002) reflection. The Bragg reflections [$(h,0,0)$ in Fig. 1] originating from the double-hcp lattice were easily observed with both neutrons and x rays, but no trace of Bragg scattering appeared elsewhere in the x-ray patterns. The dashed lines in Fig. 1(b) indicate the x-ray scattering to be expected if the $(h\pm q,0,0)$ Bragg reflections observed by

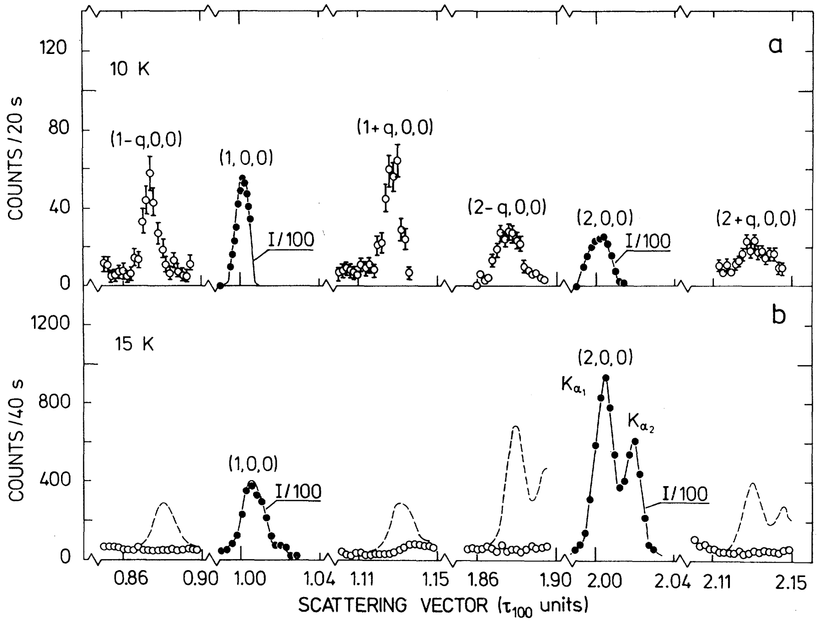

FIG. 1. (a) Neutron and (b) x-ray diffraction patterns obtained from a single-crystal neodymium plate. The scan direction was along [100] with (a) the [1$\bar{2}$0] axis and (b) the [001] axis perpendicular to the scattering plane. The dashed lines in (b) show the x-ray intensity to be expected if the $(h+q,0,0)$ satellites that are observed with neutrons (a) originate from lattice distortions. An x-ray pattern obtained with the [1$\bar{2}$0] axis vertical leads to the same conclusion as the data shown in (b).

neutrons [Fig. 1(b)] did indeed originate from lattice distortions. Thus the model interpreting the $(h\pm q,0,0)$ reflections observed by neutrons as arising from lattice distortions is incorrect and these reflections must be of magnetic origin. A recent experiment by Moon and Koehler[5] using neutron-polarization analysis is in accordance with this conclusion.

As mentioned, the above conclusion led to additional collection of neturon diffraction data which showed that the magnetic Bragg reflections are split.[6] Hence the modulation wave vector $\vec{q}$ describing the magnetic structure of neodymium has components both parallel (q_x) and perpendicular (q_y) to the $\langle 100\rangle$ axes. Figure 2 shows q_y scans at fixed q_x, with $q_x=0$, 0.11, 0.12, 0.125, 0.13, and 0.135. The split peaks observed for $q_x \neq 0$ show the scattering caused by the satellite pair at A (see the inset of Fig. 2) when smeared by the finite resolution of the spectrometer. Similar peaks were also observed around the A', B, B', C, and C' lattice points. This means that instead of giving rise to six magnetic satellite reflections around each double-hcp lattice point as previously assumed, the magnetic ordering results in twelve magnetic satellite reflections around each lattice point. The positions of these satellite reflections are illustrated by the large and small filled circles shown in the inset of Fig. 2. The intensities observed in the reflections B, B', C, and C' are larger than the intensities observed in the reflection A and A' by a factor of 10. The splitting of the satellite reflections was not observed previously because the data reported by Bak and Lebech were obtained in the same way as the data shown in Fig. 1(a), i.e., by scanning along the [100] direction through the saddle points of the split peaks shown in Fig. 2. A search around the (1,0,0) Bragg reflection for higher-order satellites turned out negative. The search was made by doing q_x scans at fixed q_y through the lattice points shown as large unfilled circles in the inset of Fig. 2.

From the data shown in Fig. 2 and similar scans around the lattice points B and $(q_x, \pm q_y, 1)$ we deduce $q_x \sim 0.128$ and $q_y \sim 0.006$ at 11.5 K. A preliminary study of the temperature dependence of $\vec{q}$ from 10 to 18.4 K suggests that q_x increases with temperature and that q_y remains almost constant up to ~15 K, whereafter it decreases. At 18.4 K we find $q_x \sim 0.14$ and $q_y \sim 0.003$ from scans through $(q_x, \pm q_y, 1)$.

It may be argued that the different temperature dependencies observed by Bak and Lebech for the

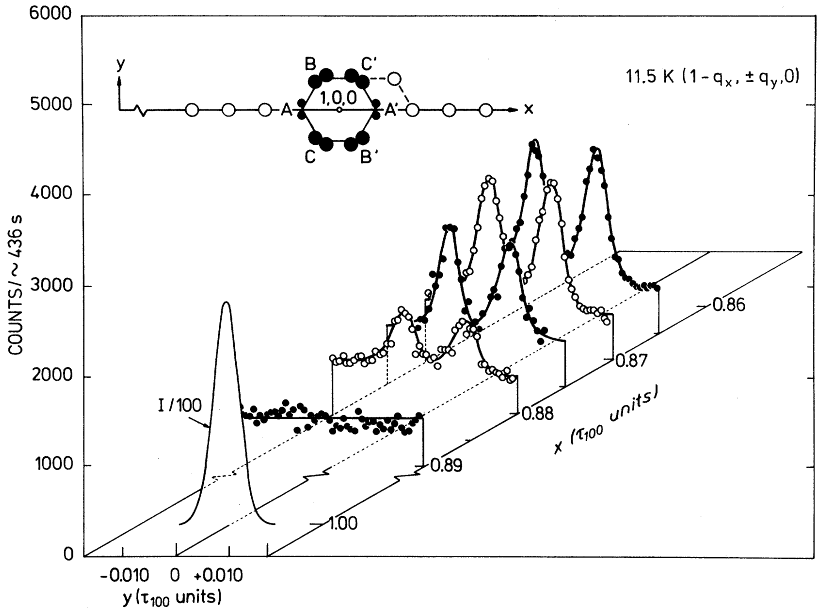

FIG. 2. Neutron-diffraction data obtained from a single-crystal neodymium plate with the [001] axis perpendicular to the scattering plane. The data were obtained using a thermal-neutron spectrometer with a poorer signal-to-background ratio than the spectrometer used to obtain the data shown in Fig. 1(a). The main figure shows q_y scans at fixed q_x, with $q_x = 0$, 0.11, 0.12, 0.125, 0.13, and 0.135 at 11.5 K. The inset illustrates the positions of the twelve magnetic satellite reflections observed around the (1, 0, 0) lattice point (filled circles) and the lattice points were higher-order satellite reflections were searched for (large unfilled circles).

magnetic reflections around (0,0,3) and (1,0,0) are evidence of a change of magnetic structure at ~18.4 K. However, this is not corroborated by other types of measurements. Neither the data on elastic stiffness constants by Lenkkeri and Palmer[7] nor the heat-capacity data by Forgan, Muirhead, Jones, and Gschneidner[8] show evidence of a magnetic phase transition between T_N and 10 K although both sets of data show anomalies at T_N and at the phase transitions observed below 10 K. Hence, we conclude that the present x-ray and neutron-diffraction studies demonstrate that the triple-$\vec{q}$ structure in the form proposed by Bak and Lebech *is not* the correct model of the magnetic structure of neodymium. The study has revealed previously unobserved magnetic Bragg reflections, which implies that the available neutron-diffraction data are incomplete. Therefore, more neutron-diffraction experiments using both unpolarized and polarized neutron scattering techniques are being planned.

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