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To cite this version:
E. Forgan, E. Gibbons, K. Mcewen. 4-q MAGNETIC STRUCTURE OF NEODYMIUM. Journal de Physique Colloques, 1988, 49 (C8), pp.C8-337-C8-338. <10.1051/jphyscol:19888150>. <jpa-00228297>

HAL Id: jpa-00228297
https://hal.archives-ouvertes.fr/jpa-00228297
Submitted on 1 Jan 1988

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4–q MAGNETIC STRUCTURE OF NEODYMIUM

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Abstract. – We have used two neutron-scattering techniques to investigate the magnetic structure of neodymium. By observations of harmonic combinations of its modulation vectors we have deduced that the low temperature magnetic structure is quadruple-q. We have confirmed this by observing a single domain of the structure in a suitably aligned magnetic field.

For more than twenty years, the magnetic ordering of neodymium has presented fascinating problems, and the behaviour of this pure crystalline element is only now becoming understood. As it is cooled below its Néel temperature of 19.9 K, dhcp neodymium undergoes a series of magnetic phase transitions [1-3] with various incommensurate wavevectors describing the ordering, until at 5 K and below, a typical Bragg point is surrounded by 36 magnetic satellites, as in figure 1. We shall describe some experiments which demonstrate that the complex low temperature pattern of satellites is the result of several domain orientations of a rather simpler structure; we shall also show one of the reasons why this structure is formed.

A close examination of figure 1 shows that it contains just four distinct wavevectors, repeated by symmetry around the basal plane. The vectors come in two similar pairs: (a) a pair \( q_1 \) and \( q_2 \) lying along the \{100\} axes (b directions) and having two different lengths \([4]\) close in value to 0.11 \( \tau_{100} \); (b) a pair \( q_3 \) and \( q_4 \) of length \( \sim 0.18 \tau_{100} \), lying at two different distances from the b-axes, giving a group of four satellites [5]. If we assume that every part of a neodymium crystal giving such a pattern is in the same energy state, then every part of the crystal must contain at least one modulation vector of each type. The simplest way for this to occur would be for the basic magnetic structure to contain only four modulation vectors; the observed pattern would then be due to the addition of the scattering from several distinct domain orientations of this basic structure in different parts of the crystal. The most complicated structure consistent with figure 1 would have 18 different modulation vectors, all present in all parts of the crystal!

In fact, strong arguments have been given against the possibility that two closely-spaced modulation vectors are present within a single region of a crystal [1]. It is therefore likely that \( q_1 \) and \( q_2 \) do not share the same b-direction, and similarly \( q_3 \) and \( q_4 \). Indeed, if the low temperature structure of neodymium is analogous to that observed between 9 and 19 K [2, 3, 6] then we would expect \( q_1 \) and \( q_2 \), and also \( q_3 \) and \( q_4 \), to form two double-q structures, each having wavevectors close to two of the three b-directions. However, even if we accept these arguments, they are insufficient to specify the magnetic structure and further information is required.

One source of information which can be obtained at low temperatures is the mixing of the ordering wavevectors to give “intermodulation harmonics”. These are due to the “squaring-up” of the sinusoidal modulation of an incommensurate structure which occurs at \( T \ll T_N \) and leads to odd harmonics, of which the third is the strongest. In a single-q structure with wavevector \( q \), the third harmonic would be at 3 \( q \). In a multiple-q structure, intermodulation 3rd harmonics can occur at any combination \( q_1 \pm q_2 \pm q_4 \). The presence of such harmonics indicates that \( q_1 \), \( q_2 \), and \( q_3 \) are present within a single domain, and the positions of the harmonics give the relative orientations of their constituent fundamental wavevectors. We emphasise that this technique may be applied in a multi-domain specimen to deduce the structure of a single domain.

We have carried out experiments on the four-circle diffractometer D10 at the Institut Laue-Langevin, Grenoble, to observe such harmonics as satellites...
around the (003) reciprocal lattice point at a temperature of 4.5 K and below. A detailed analysis of the results, which will be published elsewhere [7], indicates that the low temperature magnetic structure of neodymium is indeed quadruple-\( q \), and consists of two double-\( q \) structures, coupled together in a specific orientation. Rather than giving further details of the harmonic technique, we will describe the result of a recent neutron scattering experiment, which confirms our conclusion rather directly. This experiment was performed on the 2-circle diffractometer D20, also at the I.L.L. We measured the magnetic satellites around the Bragg point (100) with a magnetic field applied in the basal plane at about 5 degrees from the (100) direction. It should be mentioned that the satellites with vectors \( q_3 \) and \( q_4 \) have not previously been observed around this Bragg point because they are extremely weak: however the high flux and low background of the instrument D20 allowed us to detect them without difficulty.

At a temperature of 2 K, a field of 4.5 T was applied and then the field was reduced to 0.4 T, in the expectation that a single orientation of a 4-\( q \) domain might be favoured. This was indeed the case: the satellites observed corresponded to just four distinct \( q \)-vectors, the orientations of which are represented in figure 2. The sample also remained almost entirely in this single domain orientation as the applied field was reduced to zero. At other applied fields and temperatures there were complex field-induced magnetic structures, which will be reported elsewhere [8].

We close with the following remark about the 4-\( q \) structure shown in figure 2. We note that within experimental error the following vector relationship holds:

\[
q_3 + q_4 - 2q_1 = 0.
\]

This in fact to be expected: it is a necessary condition for the existence of a non-zero fourth-order term in the free energy, which will couple together the modulations with these three \( q \)-vectors.

Acknowledgments

We acknowledge the financial support of the Science and Engineering Research Council for this work, also that of the I.L.L. for a studentship for E.P.G. We thank D. Fort of the Materials Science Unit, Department of Metallurgy, University of Birmingham for his work in the preparation of single crystals, and many staff at the Institut Laue-Langevin for their advice and assistance.

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[5] This fourfold splitting has not previously been reported in the literature-only the twofold splitting of reference [4]. However it has been observed both by us and previously by B. Lebech (private communication).

