Does the "Triple-q" Magnetic Structure Exist in Neodymium?

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Through neutron polarization-analysis experiments at 9.9 K, we have determined that certain satellites in the neutron diffraction pattern of Nd are not caused by a lattice distortion, but rather by a modulated moment component in the basal plane and perpendicular to $\vec{b}_1$. The magnetic structure is thus even more complicated than hitherto proposed, and there is now no direct experimental evidence to support the "triple-$\vec{q}$" model.

In the first neutron diffraction study of Nd, Moon, Cable, and Koehler[1] discovered a complex diffraction pattern indicating modulated moment behavior and at least two magnetic phases. They proposed approximate models to account for the major features of the diffraction patterns but acknowledged that the models were inadequate to explain all the features. In particular, small satellites found at the reciprocal lattice positions $(h \pm q, 0, 0)$ in the high-temperature phase (7.5–19 K) were not explained by their model. Recently, Bak and Lebech[2] have proposed a much more complicated model for the high-temperature phase based on Landau symmetry arguments and renormalization-group theory. By combining the experimental observation that the high-temperature transition is of second order with their theoretical considerations, they reach the interesting conclusion that the stable phase must have a "triple-$\vec{q}$" structure.

To understand the meaning of this terminology, let us consider the diffraction pattern in a little more detail. The crystal structure of Nd is double hexagonal close packed. In reciprocal space the magnetic satellites are found as hexagonal arrays around reciprocal lattice points, displaced from these points by $\pm q\, \vec{b}_i$, where $\vec{b}_i$ are reciprocal lattice vectors along the three equivalent ⟨100⟩ directions.[3] In a "single-$\vec{q}$" model, three domains are required. Within each domain there is a periodic modulation along one of the three equivalent propagation directions. In a triple-$\vec{q}$ model, there is a single domain with periodic modulations along all three of the equivalent directions. Unfortunately, a single-$\vec{q}$ model with equally populated domains will give exactly the same set of intensities as a triple-$\vec{q}$ model.

The model of the magnetic structure by Bak and Lebech can be considered either in a single-$\vec{q}$ or triple-$\vec{q}$ version. In neither case does this model predict magnetic satellites at the positions $(h \pm q, 0, 0)$. By considering the spin-lattice coupling, Bak and Lebech argued that there should be a periodic modulation of the lattice with the same periodicity shown by the magnetic structure. However, this coupling involves two different components of the order parameter and so is active only in the triple-$\vec{q}$ state. A triple-$\vec{q}$ state with significantly strong spin-lattice coupling should then produce nuclear scattering intensities which superimpose on the magnetic satellites. In particular, there should be intensity of nuclear origin at the troublesome $(h \pm q, 0, 0)$ positions. The existence of these satellites was then offered as proof of the existence of the triple-$\vec{q}$ state. It should be noted that the conclusion that these satellites are caused by a lattice distortion has already been placed in jeopardy by x-ray diffraction experiments[4] in which these satellites were not found.

Through the technique of neutron polarization analysis[5] it is possible to distinguish unambiguously between nuclear and magnetic scattering and we have performed such an experiment on the $(1-q, 0, 0)$ satellite. In this experimental technique we can measure separately those neutrons which have reversed their spin on scattering and those which have maintained the same spin direction. Coherent nuclear scattering will always be spin-nonflip scattering, while magnetic scattering can be either spin flip, spin nonflip, or a mixture of the two, depending on the orientation of the electronic moments relative to the neutron polarization direction.

In our experiment we have measured the two partial cross sections for the satellite at $(\frac{7}{8},0,0)$ in two different crystal orientations related by a 90° rotation about the scattering vector. The geometry of the experiment is indicated in Fig. 1. If the scattering is nuclear in origin we expect to find that

$$N_1^{++} = N_2^{++} = N_{\rm nuc}, \tag{1}$$

and

$$N_1^{-+} = N_2^{-+} = 0, \tag{2}$$

where N_m^{ij} is the neutron intensity in orientation m corresponding to scattering from the initial neutron spin state i to the final state j. On the other hand, if the scattering is produced by a set of oriented moments which have a projection in the plane perpendicular to the scattering vector along the direction defined by $\vec{\mu}_\perp$, we expect to find that

$$N_1^{++} = N_2^{-+} = N_{\rm mag} \sin^2\beta, \tag{3}$$

and

$$N_1^{-+} = N_2^{++} = N_{\rm mag} \cos^2\beta, \tag{4}$$

where β is the angle between $\vec{\mu}_\perp$ and [001]. We have also considered the possibility that these satellites, which are quite weak, are entirely spurious, arising from double Bragg scattering. In this case there should be no relationship between the intensities in the two orientations. The intensities should have erratic behavior as a function of azimuthal angle and as a function of neutron wavelength. No easy generalization can be made about the distribution of scattering between N^{++} and N^{-+} in the double-Bragg-scattering case, it is probable that there would be a mixed distribution for orientation (1) and N^{-+} would be dominant for orientation (2).

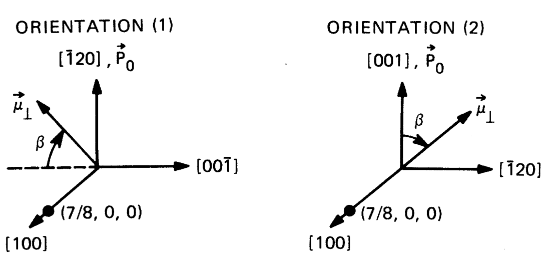

FIG. 1. Geometry of the polarization-analysis experiments. $\vec{P}_0$ shows the direction of the neutron polarization.

The experiment was performed at a temperature of 9.9 K in an applied magnetic field of 1.2 kOe. This weak field was used to control the neutron polarization without disturbing the magnetic structure of the sample. No change in the total satellite intensity was detected when the field was reduced to zero. The results are displayed in Fig. 2. The significance of the two peaks seen in Fig. 2(b) is discussed in the next paragraph. For the moment, let us integrate over both peaks in Fig. 2(b) in comparing the intensities found for the two orientations. In a good approximation, the "flipper off" intensity corresponds to N^{++} and the "flipper on" corresponds to N^{-+}. It is immediately apparent that Eqs. (1) and (2) are not valid and hence that the satellite is not due to nuclear scattering caused by a periodic lattice distortion. The small "flipper on" intensity seen in Fig. 2(a) can be explained by imperfect beam polarization [the ratio of off-on intensity for the (100) nuclear peak was 44.1 ± 1.8]. The scattering thus goes from completely spin nonflip in orientation (1) to completely spin flip in orientation (2), which is consistent with Eqs. (3) and (4) if $\beta = 90°$. When the integrated intensities of Fig. 2 are normalized to the respective (100) intensities in the

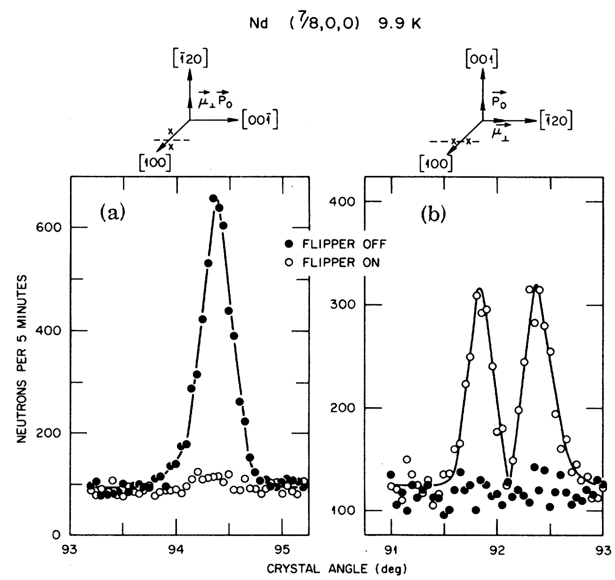

FIG. 2. Polarization-analysis results for Nd in two crystal orientations. The dashed lines in the upper diagrams indicate the direction of the spectrometer scans in reciprocal space. The crosses indicate the position of the satellites.

two orientations it is found that Eq. (3) is quantitatively satisfied with $\beta = 90°$. If we define

$$R_m^{ij} = \frac{\int N_m^{ij}(\frac{7}{8}, 0, 0)\, d\theta}{\int N_m^{++}(1, 0, 0)\, d\theta}, \quad (5)$$

we find that $R_1^{++} = (1.14 \pm 0.03) \times 10^{-2}$ and $R_2^{-+} = (1.22 \pm 0.06) \times 10^{-2}$.

To check further on the double-Bragg-scattering possibility we measured a similar ratio in orientation (2), without the analyzer crystal, for six different neutron wavelengths equally spaced between 1.0 and 1.25 Å. In this experiment we measure the sum of the (++) and (−+) intensities. We found the ratio to be independent of wavelength with an average value of $(1.29 \pm 0.04) \times 10^{-2}$. Our conclusion is that these satellites are purely magnetic in origin, arising from modulated moments which are oriented perpendicular to both [100] and [001]. The Bak-Lebech model does not contain such a feature.

The other interesting fact displayed in Fig. 2 is that we should have been taking about two satellites symmetrically displaced off the $[h00]$ axis by a small amount. This splitting has been observed previously by Lebech, Als-Nielsen, and McEwen.[4] If $\hat{a}$ is a unit vector in reciprocal space normal to both [100] and [001], then the propagation vector of the satellites we have been discussing is

$$\vec{q} = \tfrac{1}{8}\vec{b}_1 \pm 0.004 |b_1| \hat{a}, \quad (6)$$

and the moment direction is along $\hat{a}$. The satellites appear as a single peak in orientation (1) [Fig. 2(a)] with their full intensity because of the poor vertical resolution of the spectrometer.

Clearly, the magnetic structure is more complicated than anyone has hitherto proposed. The newly identified magnetic component with moments along $\hat{a}$ will produce intensities which superimpose on the regular satellite intensities when the crystal is in orientation (1), which is the richest orientation in terms of available peaks. Previous work will have to be reinterpreted in terms of a more complex model. It is anticipated that the polarization analysis technique will play an important role because in orientation (1) the $\hat{a}$ component produces spin-nonflip scattering while components perpendicular to $\hat{a}$ produce spin-flip scattering.

While we have shown that the physical evidence adduced to support the triple-$\vec{q}$ model is invalid, we have not proven that this state does not exist. The lattice distortion could be too small to be detected. The different temperature dependences reported by Bak and Lebech for the $(q, 0, 3)$ and $(1-q, 0, 0)$ satellites, which they interpreted as an indication of triple-$\vec{q}$ behavior, should probably be reinterpreted as an indication of two phase transitions separated by about 1.5 K. Their theory would then be applicable only in this narrow temperature range above the lower of the two transitions. The problem of producing evidence to support or refute the triple-$\vec{q}$ structure is formidable. If a crystal could be found which shows unequal intensities for equivalent reflections, the case for a single-$\vec{q}$, multiple-domain model would be strengthened. Such a crystal might be produced by cooling through the highest transition in a magnetic field.

Two of us would like to take this opportunity to correct a mistake made in the preparation of Ref. 1. The propagation vector of the new satellites which appear below 7.5 K is $0.185\vec{b}_1$, not $0.15\vec{b}_1$.

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

[2]Per Bak and Bente Lebech, Phys. Rev. Lett. 40, 800 (1978).

[3]In this paper $[hkl]$ refers to a direction in reciprocal space.

[4]B. Lebech, J. Als-Nielsen, and K. A. McEwen, following Letter [Phys. Rev. Lett. 43, 65 (1979)].

[5]R. M. Moon, T. Riste, and W. C. Koehler, Phys. Rev. 181, 920 (1969).