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J. Phys.: Condens. Matter **19** (2007) 286202 (12pp) doi:10.1088/0953-8984/19/28/286202

Magnetic distorted spiral structure of Nd at 10 K

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Received 26 March 2007, in final form 18 May 2007
Published 26 June 2007
Online at stacks.iop.org/JPhysCM/19/286202

Abstract
Nd has been suggested to have the unusual $2q$ sinusoidal structure below $T_N = 19.9$ K. However, here will be presented an analysis of neutron scattering data, which shows that a flat distorted spiral with aspect ratio 1:3 at $T_N/2 = 10$ K provides a better fit and a physically more appealing model. It turns into a striped (sinusoidal) structure only at $T_2 = 19.1$ K, very close to T_N, where the non-magnetic stripes are formed. A simple mean field theory demonstrates that this transition may be driven by a minute anisotropy. A model of randomly placed singlet pairs á la Anderson resonance-valence-bond (RVB) phase is discussed. The observed splitting up of satellite peaks below T_2 is discussed in terms of an epitaxial rotation model. The results are discussed in relation to previous models.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Nd has the most complicated magnetic structure of any pure element [1]. The lattice structure is dhcp with sites of nearly cubic symmetry (A) alternating with sites of nearly hexagonal symmetry (B, C) as ABACA. In the first study [2], Nd was suggested to assume an antiferromagnetic, incommensurate sinusoidal order on the hexagonal sites (only) at the ordering temperature, $T_N = 19.9$ K. Nd was the first material for which such an unusual order was suggested. Below $T_2 = 19.1$ K additional complexity sets in. After a number of further studies [3–5], the following model was proposed for $19.1 > T > 9$ K. The structure should consist of sinusoidal ordering on the hexagonal sites with an ordering vector $\mathbf{Q} = (001) + \boldsymbol{\delta}$, where the satellite vector $\boldsymbol{\delta}$ is close to (canted up to $\pm 2.5°$ away from the b-direction [5]) any of the six equivalent vectors $\boldsymbol{\delta} = \langle \delta 00 \rangle$, say $\mathbf{Q} = (\delta, 0, 1)$ r.l.u., where $\delta \sim 1/7$, and it is temperature dependent. The moment on the hexagonal sites should be in the basal (ab-) plane and canted [5] up to $\pm 15°$ away from the b-direction in the dhcp lattice structure. We use here Cartesian coordinates $(a, b) = (x, y)$. In addition, sinusoidal magnetic order should be induced on the cubic sites with the major component along the c-axis. The physical basis for this exceptional structure has been a puzzle for almost half a century and inspired numerous studies [2–12].

The $1q$ sinusoidal ordering implies a variable moment size on each site. This is energetically unfavourable versus the normal spiral ordering found in other rare earth [1] materials (Tb, Dy). Further it has been difficult to find an origin for a strong anisotropy, which would appear necessary to favour such a longitudinal order—and why is it canted up to $\pm 15°$ away from the symmetry direction? This was rationalized by Forgan [12], who suggest it was a $2q$ canted, sinusoidal structure—based on a Landau expansion. The spiral structure avoids the problem with the variable size moments and also with the moment direction. The average magnetic interaction energy is given by $E_{\text{spiral}} = -J(\mathbf{Q})(\langle M^x_{\mathbf{Q}} M^x_{-\mathbf{Q}}\rangle + \langle M^y_{\mathbf{Q}} M^y_{-\mathbf{Q}}\rangle)$, where $J(\mathbf{Q})$ is the Fourier transformed (FT) interaction function, with a maximum at any of the equivalent $\mathbf{Q}$ vectors, $M^x_{\mathbf{Q}}$ is the FT moment component along $\mathbf{Q}$, and $M^y_{\mathbf{Q}}$ the FT transverse component. A flat spiral has $M^x_{\mathbf{Q}} > M^y_{\mathbf{Q}}$, but the moment increases when lowering the temperature; it is favourable to increase the components towards the same length. The $1q$ sinusoidal structure clearly has only the first term; and it has in fact the same smaller energy even if the moments are canted away from the $\mathbf{Q}$-direction: it is simply missing the term from the transverse component. However, if we have two simultaneous ordering vectors with moments canted oppositely from the equivalent $\mathbf{Q}_1$ and $\mathbf{Q}_2$ vectors, we will get the average energy $E_{2q-\text{sinusoidal}} = -J(\mathbf{Q}_1)\langle M^1_{Q1} M^1_{-Q1}\rangle - J(\mathbf{Q}_2)\langle M^2_{Q2} M^2_{-Q2}\rangle$. Since $J(\mathbf{Q}_1) = J(\mathbf{Q}_2)$, we then get exactly the spiral energy when the M^1_{Q1} is perpendicular to $M^2_{Q^2}$ and of equal length. This happens for the canting of $\pm 15°$ in a hexagonal structure. Lowering the temperature should therefore make the canting approach that to gain transverse energy. This is basically the physics behind Forgan's model. The transverse energy there is simply added using another ordering vector $\mathbf{Q}_2$ with another moment direction. In the spiral structure it is always included, whether it is of the single-q or the multi-q kind. Neither a neutron nor an x-ray scattering experiment can distinguish between a multi-domain single-q structure and a one-domain multi-q structure (without special treatment by fields or strain etc). However, as we shall see, experiments can distinguish between the spiral and the $2q$ sinusoidal models.

Neutrons measure the bulk of a crystal, and therefore average over many domains. The intensity depends on a polarization factor $(1 - (\boldsymbol{\kappa} \cdot \mathbf{m})^2)$, where $\boldsymbol{\kappa}$ and $\mathbf{m}$ are the unit scattering and moment vectors, respectively. It was on the basis of the dependence on this factor alone that the sinusoidal structure was proposed [2]. Magnetic x-ray scattering probes a relatively small region near the surface and may therefore measure only one or a few domains. It also has a different polarization factor. The techniques are therefore complementary. A multitude of satellite peaks has been observed by neutron scattering around the lattice Bragg peaks, and the corresponding simple antiferromagnetic Bragg peaks. The $1q$ sinusoidal structure on the hexagonal sites cannot explain an apparent erratic asymmetry in the satellite peak intensities around these Bragg peaks, where full symmetry is generally expected. This problem was pointed out in the very first neutron study by Moon *et al* [2]. A polarization of the cubic sites appears necessary. Below $T_2 = 19.1$ K a small splitting up of the satellites was later observed [5], where $\boldsymbol{\delta}$ turns by up to $\pm 2.5°$ away from the b-direction; further, a transverse moment component (in the a-direction) is needed. There is no further phase transition between T_2 and $T_1 \sim 9$ K, where the cubic sites also order. Here we shall demonstrate the magnetic structure of Nd in this temperature interval can equally well (or slightly better than previously) be described as a flat spiral order on the hexagonal sites, inducing a similar spiral order on the cubic sites, but with no c-axis component. This simple model is clearly more physically appealing. It might be a $2q$ structure; the present data cannot tell. Multi-q structures and previous models for the Nd structure will be discussed further in the appendix.

The possibility of a sinusoidal, striped structure has been investigated for the (HTc) high temperature superconducting oxides [13, 14]. In fact there is a surprising similarity in the magnetic properties between these and Nd. Both are essentially planar structures with weak interactions between planes, having a nearly degenerate interaction energy surface [15, 16] and strong magnetic disorder. A better understanding of the order in Nd may be instructive

Table 1. Integrated neutron intensities [17] in arbitrary units for the magnetic satellites in Nd at $T = 10$ K. The data have been corrected for the Nd form factor [4]. Notice the approximate symmetry around $h = 0$ (the deviation from which probably indicates the experimental accuracy), but notice in particular the large asymmetries between the $\tau \pm \delta$ satellites. Data not indicated are assumed not to be measured.

$\ell \backslash h$	$-1-\delta$	$-1+\delta$	$0-\delta$	$0+\delta$	$1-\delta$	$1+\delta$	$2-\delta$	$2+\delta$
0	2.8	2.4			2.2	2.8	5.3	
1	6.7	1.0	30.3		1.1	6.5	2.3	5.6
2	11.4	12.1			11.8	10.6	4.4	6.1
3	2.2	12.8	55.0		11.5	2.3	9.6	
4	19.3	31.6			27.1	15.0	6.5	10.1
5	18.5	4.7		45.2	3.5	15.2		11.3
6	49.5				20.5	43.8	17.7	11.6[a]
7	4.9	14.5		49.6	12.6	6.0	14.1	
8		44.2			29.6	19.6	15.7	24.7
9			49.1	51.1	5.9	16.5		
10						32.9		
11						10.1		

[a] The intensity given at $(2+\delta, 6)$ was in error in the original table, and has here been corrected [4].

for understanding the even more complex properties of the HTc materials, where neutron scattering measurements are extremely difficult because of the small moment ($S = 1/2$). Here we concentrate on understanding the Nd structure.

2. Experimental data and fit to the flat spiral model

An extensive neutron scattering study at $T = 10$ K (just above the cubic ordering temperature $T_1 \sim 9$ K) of 58 magnetic intensities in the $(h, 0, \ell) \equiv (H, L)$ plane was made by Hansen [17], table 1, but no analysis was provided. However, the data contain the key to understanding the complicated Nd magnetic structure. Later, Lebech [3] discussed a fit to the data using the above canted, sinusoidal model with a cubic c-axis component. She pointed out that a full understanding was not achieved. This fit, together with the fit to x-ray data with a similar model by Watson *et al* [11], will be discussed in the appendix. Here I shall as a first step use a simplified model, and show it fits all the observed intensities satisfactorily by means of essentially only four parameters. As ordering vector for the hexagonal sites we assume $\mathbf{Q} = (001) + \boldsymbol{\delta}$, where $\boldsymbol{\delta} = (\delta 00)$. For simplicity we take $\delta = 1/7$ for both hexagonal and cubic sites (since the cubic ordering is induced [18] by that on the hexagonal sites). In general $\boldsymbol{\delta} = \langle \delta 00 \rangle$ is any of the equivalent vectors in the sixfold star. We neglect that these ordering vectors are not perfectly along the $\langle 100 \rangle$ directions—it is not important for the intensity calculation; neither is it important whether the structure is of a single-q or multi-q type. The result is that it *need not be* a sinusoidally ordered structure in the temperature range $10\text{ K} < T < 19.1$ K. Instead, it may be described by a flat spiral order of the spins on the hexagonal sites n in planes 1 and 3, which are antiferromagnetically ordered between the planes with a possible, additional phase shift φ. Further, there is a weak polarization of the spins on the intervening cubic layers in planes 2 and 4 with a phase shift ψ. The model is

$$\begin{aligned} M_1^\alpha(n) &= M_{\text{hex}}^\alpha F^\alpha(2\pi\delta(n+\rho)+\chi) \\ M_3^\alpha(n) &= -M_{\text{hex}}^\alpha F^\alpha(2\pi\delta(n+2\rho)+\varphi+\chi) \\ M_2^\alpha(n) &= M_{\text{cub}}^\alpha F^\alpha(2\pi\delta n+\psi+\chi) \\ M_4^\alpha(n) &= M_{\text{cub}}^\alpha F^\alpha(2\pi\delta n+\psi+\chi), \end{aligned} \qquad (1)$$

where $\alpha = x$ is in the hexagonal b-direction and $\alpha = y$ is in the a-direction, and $F^x(\Omega) = \cos(\Omega)$, $F^y(\Omega) = \sin(\Omega)$, $\rho = 1/3$. The overall phase χ cannot be determined in the scattering experiment, but may for commensurate structures be fixed by the hexagonal anisotropy.

The hexagonal planes are antiferromagnetically coupled, whereas the cubic planes are here assumed to be ferromagnetically coupled. An alternative model, similar to that proposed by Moon *et al* [2], in which the moments on the cubic planes 2, 4 are also antiferromagnetically ordered, yield as well a reasonably good fit to the data. Qualitatively, this model predicts no intensity at ($H = 0$, L even) and asymmetry of the ($H = 0$, L odd) satellites, whereas the model discussed here predicts full symmetry of the intensities around ($H = 0$, L odd). Inclusion of the phase φ can provide intensity at the ($00L$ even) satellites. Very small such intensities are reported for the x-ray experiment [11]; some intensity was also observed in the neutron study [4], but judged too small and uncertain to be reported. These aspects of the models can hence be distinguished by experiments, but not by the present data. Corrections for the Nd form factor have been included in the data set [4]. If we assume there should be symmetry in the data around the $H = 0$ axis, we can from the scatter estimate the experimental accuracy on the points to be about $\pm 10\%$. The average intensity of a pair of satellites is clearly more accurate than the difference intensity. The $H = 1$ data are by far the most complete, and thus the most accurate, whereas the difference intensities for $H = 2$ are the most uncertain, probably at least $\pm 20\%$. The present model differs further from the one proposed by Moon *et al* [2] by including a significant, modulated y-component, which makes the structure a flat spiral rather than a sinusoidal structure. Such a structure was already suggested to be likely, based on an analysis of the neighbouring element Pr [19]. The resulting parameters are the average moment amplitude components on the hexagonal and cubic planes (in arbitrary units μ, to be determined):

$$\begin{aligned} M^x_{\rm hex} &= (0.97 \pm 0.02)\mu, \\ M^y_{\rm hex} &= (0.29 \pm 0.01)\mu, \\ M^x_{\rm cub} &= (0.21 \pm 0.02)\mu, \\ M^y_{\rm cub} &= (-0.07 \pm 0.01)\mu, \end{aligned} \tag{2}$$

assuming $\phi = 0$ and $\psi = \pi/2$. Figure 1 shows the resulting fit to the average intensity $I_{\rm av}(\boldsymbol{\tau}) = [I(\boldsymbol{\tau} - \boldsymbol{\delta}) + I(\boldsymbol{\tau} + \boldsymbol{\delta})]/2$ and difference intensity $I_{\rm diff}(\boldsymbol{\tau}) = [I(\boldsymbol{\tau} - \boldsymbol{\delta}) - I(\boldsymbol{\tau} + \boldsymbol{\delta})]/2$, where the satellites have been measured around a Bragg point $\boldsymbol{\tau}$. In view of the experimental accuracy a more detailed fit with more parameters is not warranted. As good a fit can be obtained yielding essentially the same four amplitude parameters when also fitting φ and ψ; they do not deviate significantly from the chosen values. A fit allowing, additionally, a finite component, $M^z_{\rm cub}$, along the c-direction has been made; $M^z_{\rm cub}$ is zero within the error bars. The analysis of the present neutron data [17] does not permit an absolute determination of the moments in $\mu_{\rm B}$, i.e. of the scale factor μ, which is just unity in the fit. However, by comparing with calculations [18] of the moment versus temperature, one finds that the moment components in $\mu_{\rm B}$ at $T = 10$ K are obtained by multiplying the above parameters by a factor $\mu = 2.5\ \mu_{\rm B}$. Certain intensity properties yield information of qualitative nature: the special behaviour at $L = 6$ can only be explained by introducing the polarization of the cubic sites. Similarly, the intensity at $L = 0$ demands the presence of the perpendicular and modulated (y) components. The overall relative magnitudes of the $H = 0$, 1 and 2 data are well given by the $[1 - (\boldsymbol{\kappa} \cdot \mathbf{m})^2] = \ell^2/(r^2h^2 + \ell^2)$ factor by assuming all moments lie in the (x, y) plane [001]; $r = a^*/c^*$ is the ratio of the reciprocal lattice constants. A section of the resulting structure for $\delta = 0.143 \sim 1/7$ of the *average* moments is shown in a projection on the [001] plane at the top in figure 2.

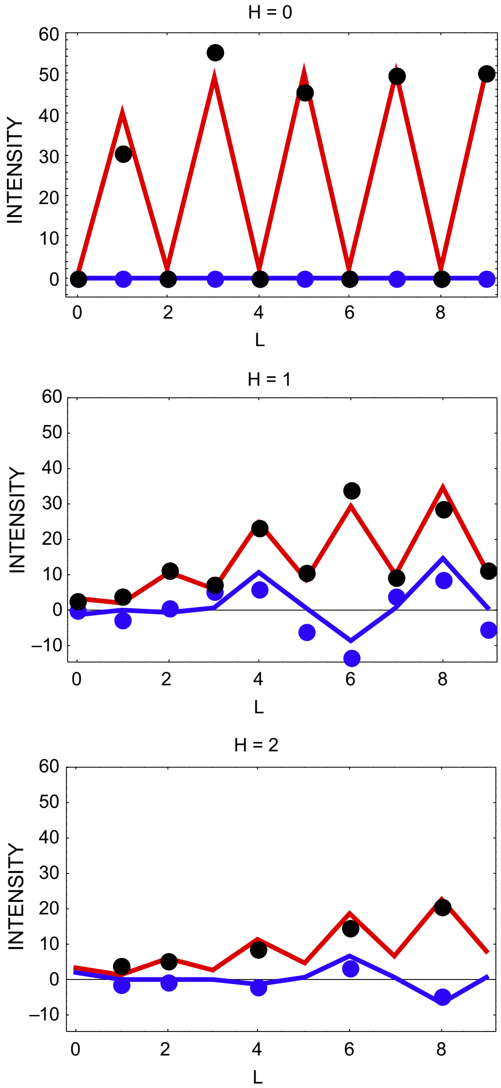

Figure 1. Data points derived from table 1 of the average and difference intensities, $I_{av}(\tau)$, black dots, and of $I_{diff}(\tau)$, blue dots. The results of the model are plotted as red and blue lines respectively for the $(h, 0, \ell)$ positions, referred to by (H, L). The lines join the calculated intensities. The data for the satellites around $h = \pm 1$ are experimentally best determined. Assuming there is symmetry around $h = 0$, the mean, where possible, is used. That is, we have used the average intensity for $H = 1 - \delta$ and $H = -1 + \delta$, as well as for $H = 1 + \delta$ and $H = -1 - \delta$. Notice that the large asymmetries at $L = 6$ are well accounted for both for $H = 1$ and 2 (check the corresponding data points, which appear quite exceptional in table 1). It is assumed that due to the symmetry the intensities of the $(-\delta, 0, \ell)$ and the $(+\delta, 0, \ell)$ are identical, hence the zero data points are also included for $I_{diff}(\tau)$, with no intensity for $H = 0$. This is in agreement with the model. The alternative model proposing antiferromagnetic ordering also of the cubic sites has finite $I_{diff}(\tau)$ for $H = 0$ and L even—and can hence be distinguished experimentally.

3. Flat spiral turning sinusoidal—possibility of a striped quantum phase

Let us discuss how this structure may arise in a Heisenberg model with strong planar and weak hexagonal crystal field anisotropy—plus a possible two-ion anisotropy of dipolar symmetry

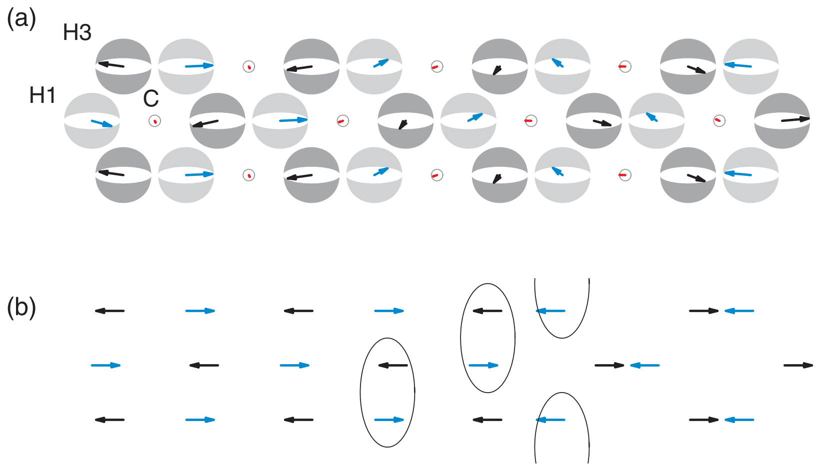

Figure 2. (a) The flat elliptic spiral structure for Nd at $T = 10$ K $\sim T_N/2$—in a projection on the [001] plane, the b-axis (x-axis) is horizontal, and the a-axis (y-axis) is vertical. The magnitude and direction of the moments are shown. The dark, H1, discs (which would be all explored in a simple spiral structure) represent the hexagonal sub-lattice 1, with (thermal average) moment directions indicated by black arrows, and the lighter, H3, discs indicate sub-lattice 3, with blue arrows. The moment stays within the indicated ellipses. The small circles, C, show the small, induced moments (red) on the cubic sub-lattices. Notice that the largest polarization is at positions where the hexagonal moments are smallest. (b) The proposed structure near T_N consists of stripes of antiferromagnetically ordered spins in the b-direction (same positions as above), separated by non-magnetic stripes of randomly placed singlet pairs, shown as the enclosed spin pairs. Only on average may this structure appear as a sinusoidal ordered state. The singlet pairs are placed where the moment components are small in the flat spiral structure—and pointing away from the b-direction. The shown spins in the 'sinusoidal' structure extend from the same centres as in the above picture. The cubic sites are not shown here.

to lowest order. Nd has angular momentum $J = 9/2$ and the full ionic moment is $g\mu_B J = 3.27\ \mu_B$. The crystal field splits the energy levels into almost pure doublets with $|\pm 1/2\rangle$ lowest. At $T = 10$ K both the $|\pm 1/2\rangle$ and $|\pm 3/2\rangle$ states are polarized [18], giving a moment on the hexagonal sites $M_{hex} = \sim 2.5\ \mu_B$. The polarization on the cubic sites was (using a full level scheme) calculated [18] to be $M_{cub} \sim 0.5\ \mu_B$, which is in good agreement with the here obtained $2.5\mu_B ({M^x_{cub}}^2 + {M^y_{cub}}^2)^{1/2} = 0.55\ \mu_B$. With moments of this size a reduction at certain sites can be explained, classically, as an increased amplitude of precession in the weaker molecular field at the node points of the magnetic structure; this is incompatible with the sixfold lattice symmetry for $\delta \sim 1/7$. Considering the molecular field from nearest neighbours in the same and opposite sublattices gives a surprising result for small $\delta \sim 1/7$. If the inter- and intra-sublattice interactions are isotropic, the magnitude of the mean field, but not direction, is identical on all sites, and a simple spiral structure with equal size moments will result. This may be perturbed by the hexagonal field, but this cannot result in an elliptic deformation. This is the case for the hcp rare earth elements (Tb, Dy) and explains the observed spiral ordering in these materials [1]. However, if the coupling is slightly anisotropic so that the longitudinal (x) and transverse (y) interactions along the satellite vector $\boldsymbol{\delta}$ are different, $J_\parallel \neq J_\perp$, the total molecular field at site n, $\mathbf{H}_{MF}(n)$, which is the sum of the interactions with the neighbouring moments, is strongly reduced at the node regions. If the anisotropy has dipolar symmetry, it will locally be uniaxial along $\boldsymbol{\delta}$, whereas globally it follows the lattice symmetry and appears of hexagonal symmetry. We can estimate the effect using the mean

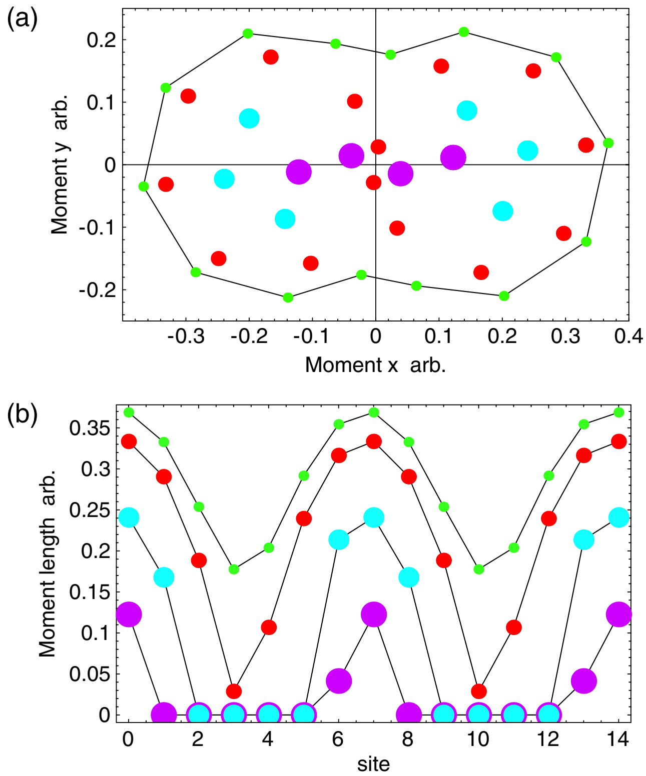

Figure 3. (a) The calculated moment length and direction for 14 consecutive H_1 and H_3 sites (belonging to planes perpendicular to the x-axis; see figure 2), assuming a small two-ion anisotropy of 4% difference: $J_\perp = 0.96J_\parallel$. The dots with increasing size represent $T = 18.9$, 19.1, 19.5 and 19.8 K (green, red, magenta, violet). It is a staggered structure, but for clarity we may invert the moments on the H_1 sites. The green dots are joined to guide the eye and the sites then follow sequentially along the line. Notice at $T = 18.9$ K (smallest, green discs, joined by lines) the overall nearly elliptical distribution. However, at the transition temperature $T_2 \approx 19.1$ K a significant part of the transverse moment vanishes (four of the second smallest, red discs are rapidly approaching zero). Notice the tendency to form the collinear, almost sinusoidal order with increasing temperature. An overall phase $\chi = 0.03\pi$ has been chosen to demonstrate this most clearly. Hexagonal anisotropy is not included for clarity, i.e. the moments are not allowed to turn towards the anisotropy-favoured x-axis. (b) The total (absolute) moment length at the same site sequence as above, joined with lines (and plotted equidistantly for simplicity). The dots represent the same as above. Notice that the non-magnetic stripe regions are formed at $T \sim 19.1$ K (second smallest, red dots).

field theory. Consider a row of sites in the b-direction, as in figure 2. Approximately, the moment for $S = 1/2$ is determined by $M(n) = \tanh(\mathbf{M}(n) \cdot \mathbf{H}_{\mathrm{MF}}(n)/kT)$, which near T_{N} yields the moment at site n as $M(n) = \surd(H_{\mathrm{MF}}(n) - kT)/H_{\mathrm{MF}}(n)$. This gives clearly an elliptic distortion, with increasing ellipticity when approaching T_{N}. At $T = 19.1$ K the regions with predominantly transverse order turn non-magnetic and form stripes perpendicular to the considered row. The loss of transverse energy may be compensated by turning the remaining spins into the b-direction, which is favourable for the anisotropy. Hence, a transition to a collinear average structure is predicted at $T_2 = 19.1$ K already with a few per cent two-ion anisotropy: $J_\perp = 0.96\ J_\parallel$. Figure 3(a) shows the thus calculated moment direction and size for $T = 18.9$, 19.1, 19.5 and 19.8 K with progressively larger dots. The display depends on the overall phase shift χ. Here a small phase shift $\chi = 0.025\pi$ is chosen, which almost yields a sinusoidal structure at $T = 19.8$ K—even without turning the spins. It requires the two-ion anisotropy and/or the hexagonal crystal field anisotropy to lock the (average) moment into the b-direction. Figure 3(b) shows the calculated moment length at various sites. It is the temperature, and the discrete lattice in combination with the long wavelength magnetic structure (given by the small, generally incommensurate satellite vector $\boldsymbol{\delta}$), which is important for creating the flat spiral structure at relatively low temperatures compared to T_N by enhancing various anisotropy effects. Close to T_N the same argument gives stripes with zero moments in the node regions and a narrower ellipse, eventually leading to the collinear, striped structure. As usual with striped structures, the assumption is that the stripes are not straight, but meander as in a liquid crystal. This finally gives the sinusoidal structure on average. Since the structure may be stabilized by a minute anisotropy, one would expect the spin fluctuations above T_N to be almost isotropic. Very interestingly, however, this is not found to be the case by neutron scattering [15]. A solution to this problem is proposed in the following.

The mean field cannot distinguish between whether the absence of average moment is due to large (paramagnetic) precessions or due to the formation of a quantum mechanical singlet state. In the first case the diffuse scattering close to T_N must show intensity due to the transverse components. This has been proved to be totally absent in a recent measurement [15] of the diffuse scattering above T_N up to $T = 25$ K. However, the transverse intensity will be absent if pairs of anti-parallel spins on opposite sublattices form singlets $(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)/\sqrt{2}$ with random partners—such that the variation of the moment only on *average* is sinusoidal. In the effective honeycomb lattice there are many ways of forming such pairs; see figure 2. The enclosed spin pairs are non-magnetic and give rise to no elastic or quasi-elastic scattering. The structure, which is indicated at the bottom of figure 2, much resembles the RVB structure proposed by Anderson [20] for triangular lattices. It has stripes of oppositely oriented antiferromagnetic order separated by non-magnetic stripes. The present $(h, 0, \ell)$ data cannot distinguish between a single-q or a multi-q structure. Many studies of Nd have been concerned with possible multi-q structures (corresponding to the checkerboard structure in the cuprates). The neighbouring Pr with $J = 4$ is also dhcp, and has a non-magnetic $|0\rangle$ ground state. It assumes, when ordered by alloying with Nd, a flat distorted spiral structure [19]. The alloys, Nd_xPr_{1-x}, are hence similar to doped cuprates, in which the Cu^{2+} atoms carry no spin, in analogy to Pr. A complete understanding of the intriguing magnetic properties of Nd and NdPr alloys is of interest in its own right—however, the implication for the elucidation of properties in the high T_c materials makes it even more important.

4. Canting of the Q-vector

In Nd the consensus is that below $T = 19.1$ K the structure becomes a $2q$ structure with satellite vectors about 120° apart forming small angles (2.5°) between the $\langle 100\rangle$ directions. This is observed as a small splitting up of the satellites observed above 19.1 K. In the accompanying paper [15] it is shown that such a splitting involves a movement of the ordering vector, i.e. the satellite vector $\boldsymbol{\delta}$, on a very flat ring of an interaction energy surface. It therefore costs little energy to change direction. Forgan [12] suggested the driving force for the canting of $\mathbf{Q}$ by 2.5° was the canting of the moment direction (up to 15°), which the $\mathbf{Q}$-vector was attempting to follow, but hindered by being caught at a pronounced peak in $J(\mathbf{Q})$. This picture is not supported by the found rather flat ring, on which $\mathbf{Q}$ can change direction with low energy cost.

Ordering off symmetry directions is also observed in the case of adsorbed monolayers of atoms on a substrate with a hexagonal surface, where there is a lattice mismatch between the adlayer and the substrate. This is called epitaxial rotation. A theory for this was developed [21] and it was demonstrated to be a case of 'ordering by disorder'. The adlayer gets distorted and the corresponding Bragg peaks develop diffuse anisotropic tails, the symmetry of which depends on the actual mechanism. When these tails overlap maximally with the substrate Bragg peaks a gain in elastic energy can be obtained. This can be achieved by a small rotation of the adlayer relative to the substrate. The epitaxial angle depends on whether it is elastically or defect driven. This effect is not active in perfect lattices with δ-function sharp Bragg peaks (only by considering effects of 'accidental coincidences' of higher order Bragg peaks [9], for which there is no evidence). For Nd, it is also essentially a planar problem in which a 2D incommensurate (and imperfect) magnetic structure must be accommodated in the hexagonal planes of the dhcp structure. Hence, the analogy is very close to the adlayer problem, as also pointed out by Lebech *et al* [9]. An analysis of the epitaxial angle may give information about whether the mechanism is driven elastically or by magnetic defects such as spin slips. The latter is most likely, as there have been observed higher order satellites [7], which signifies a squaring-up of the spin structures. On the other hand, the epitaxial rotation angle of 2.5° is small. For the adlayer case the elasticity driven rotation generates smaller angles than the defect driven one.

At low temperatures, where the hexagonal anisotropy plays a larger role, the flat spiral may be further distorted. Having the moments bunch around easy directions—or making spin slips—may lower the anisotropy energy. The present experimental accuracy does not allow us to study such structure refinements.

5. Conclusion

In conclusion, based on measured, extensive elastic neutron scattering intensities, it has been demonstrated that the magnetic order on the hexagonal sites in Nd at $T = 10$ K, and in the temperature range $9 \text{ K} < T < 19.1$ K, is well described as a flat spiral structure with aspect ratio 1:3. This induces a similar spiral order on the cubic sites. This model fits the data equally well or better than the previous $2q$ canted sinusoidal model. The small splitting of the satellites off the b-direction has been explained in terms of an epitaxial rotation model. The transition from the flat spiral to the sinusoidal phase, very close to T_N, is discussed, and it is shown that it can be stabilized by a very small in-plane anisotropy. A possible state with non-magnetic stripes consisting of singlet pairs is discussed.

Acknowledgment

It is a pleasure to acknowledge discussions with B Lebech.

Appendix. Comments on multi-q structures and models of the magnetic ordering in Nd

The structure of Nd has been a puzzle for half a century. It is the first system proposed to possess the unusual sinusoidal structure. However, most of the discussion lately has been around possible multi-q structures. This has unfortunately overshadowed the basic problem of understanding the nature of a possible sinusoidal structure. There seems to be some confusion around multi-q structures in a number of magnetic systems. Let us therefore first discuss what is, and what is not, a multi-q structure, and when can it be determined by a scattering experiment with neutrons or x-rays. We shall consider three cases.

(1) The moment $\mathbf{M}(\mathbf{r})$ at site $\mathbf{r}$ is modulated by a wave described by several Cartesian coordinates: $\mathbf{M}(\mathbf{r}) = \boldsymbol{\mu} \cos(\mathbf{Q} \cdot \mathbf{r})$; where $\mathbf{Q} = (q_x, q_y, q_z)$, where in general $q_x \neq 0$, $q_y \neq 0$, $q_z \neq 0$ and $\boldsymbol{\mu}$ is a vector which assumes some angle with respect to $\mathbf{Q}$. This is a

common situation in magnetism, and it is not a multi-q case. For example, a Nd single-q structure is described by $\mathbf{Q} = (\delta, 0, 0) + (0, 0, 1)$, where $\delta \sim 1/7$ r.l.u. This case can be distinguished in a scattering experiment with characteristic (satellite) Bragg peaks.

(2) Various moment components are modulated by a $\mathbf{Q}$ vector as above (which is one out of several equivalent, symmetry related $\mathbf{Q}_n$ vectors), but with a phase difference:

$$\mathbf{M}(\mathbf{r}) = (\mu_x \cos(\mathbf{Q}\cdot\mathbf{r}), \mu_y \cos(\mathbf{Q}\cdot\mathbf{r} + \theta_y), \mu_z \cos(\mathbf{Q}\cdot\mathbf{r} + \theta_z)).$$

For simplicity, take $\mu_z = 0$. If we assume $\theta_y = 0$, we have a *canted sinusoidal* structure with, in general, variable size moments at all sites. However, for $\theta_y = \pi/2$ we get a *spiral* structure with the same moment on all sites for $\mu_x = \mu_y$, and a *flat spiral* if $\mu_x \neq \mu_y$. Scattering experiments are very sensitive to the relative phases, and can thus, in principle, distinguish between a sinusoidal and a spiral structure. This is not a multi-q case either.

(3) Finally, the moment is locally modulated by two or several equivalent ordering vectors, say $\mathbf{Q}_1 \neq \mathbf{Q}_2$, where the moments $\boldsymbol{\mu}_i$ are related to $\mathbf{Q}_i$ with a specific angle: $\mathbf{M}(\mathbf{r}) = \boldsymbol{\mu}_1 \cos(\mathbf{Q}_1\cdot\mathbf{r}) + \boldsymbol{\mu}_2 \cos(\mathbf{Q}_2\cdot\mathbf{r} + \theta)$.
This is the proper multi-q (here $2q$) case. The scattering will show satellites at two (four) different places in reciprocal space, which must be analysed separately. One would for *symmetry reasons* expect that the amplitudes (and thus the intensities) should be equal, i.e. $|\boldsymbol{\mu}_1| = |\boldsymbol{\mu}_2|$. Likewise, one would expect the angle between $\boldsymbol{\mu}_i$ and $\mathbf{Q}_i$ to be the same (or at least symmetry related) for $i = 1, 2$. Indistinguishable from this in either neutron or x-ray experiments is if the scattering at two equivalent satellite positions arises from scattering from a population of corresponding single-q domains. For *statistical reasons* one would expect the populations to be equal. However, one would not expect to observe the same intensity under all circumstances (i.e. under external strain, external magnetic field or near a surface). A better proof of the multi-q structure is given by finding higher order satellites [7]. These can hardly arise in a multi-domain case. For Nd as example, we may take $\mathbf{Q}_1 \sim (\delta, 0, 0) + (0, 0, 1)$ and $\mathbf{Q}_2 \sim (0, \delta, 0) + (0, 0, 1)$ including the observed few degree canting from the symmetry directions.

In this light we shall discuss the previous models for the magnetic structure of Nd. Lebech [3] made a fit to the present data by assuming a $1q$ canted sinusoidal structure and including moments on the cubic sites—in particular a component along the c-axis. The parameters (scaled to optimally fit the average intensities) are given in table A.1. Watson *et al* [11] measured the Nd magnetic structure using x-rays (the penetration dept is at the L-edge of the order of 10 μm [22]). They analysed their data with a similar canted sinusoidal model, but included also (in the present notation) two different small phases on the hexagonal planes. Both turned out to be very small, $\sim 1°$. In the present notation this corresponds to assuming $\psi = \pi/2 - 1°$ on the cubic planes. It is remarkable that a fit to the 84 x-ray data points gives parameters very close to those found by Lebech [3]. The deduced parameters (translated into the notation of this paper, and choosing phases to match signs) are given in table A.1. Again a significant component along the c-axis on the cubic sites is needed. They give actually a better fit than the Lebech parameter set. There is almost no change if we set $\psi = \pi/2$, so both models use essentially five parameters. Does this mean Nd has a sinusoidal structure at $T \sim 10$ K, with a significant moment component on the cubic sites along the c-axis? It might be so, but in the first part of this paper we demonstrated that an even slightly better fit can be obtained using the flat spiral structure with just four parameters.

Let us now compare and discuss the three fits or models. The resulting average intensities of satellite pairs are indistinguishable and within the line width of the plots in figure 1, although not identical. The difference intensities show some differences. The Lebech model gives a marginally better fit to the $H = 1$ data than the two others, but a worse fit to the $H = 2$

Table A.1. Comparison between the present four parameter flat spiral model, the five parameter sinusoidal model by Lebech [3], and the seven parameter sinusoidal model by Watson *et al* [11] fitted to a completely different dataset obtained by x-ray scattering. The latter gives a set of calculated intensities which are essentially indistinguishable from those shown in figure 1, with minor differences for $H = 2$. So does the Lebech fit except for being less successful fitting the $H = 2$ data. The sum over the squared deviations is indicated.

	Spiral model present (4 par)	Sinusoidal model Lebech [3] (5 par)	Sinusoidal model Watson *et al* [11] (7 par)
M^x_{hex}	0.97 (0.02)	0.81	0.82
M^y_{hex}	0.29 (0.01)	0.19	0.19
M^x_{cub}	0.21 (0.02)	0.09	0.05
M^y_{cub}	−0.07(0.01)	0.016	0.01
M^z_{cub}	0	0.20	0.13
φ	0	0	0
ψ	$\pi/2$	$\pi/2$	$\pi/2 - 1°$
$\Sigma_i(\Delta_i^2)/N$	9.9	23	12.8

data (this might be because the $H = 2$ data have been weighted low due to the experimental uncertainty). The difference intensities are indistinguishable for the flat spiral model and the sinusoidal Watson *et al* [11] model from the x-ray data set, and the fit is as shown in figure 1. Conversely this means that the spiral model can presumably also fit the x-ray data. The various model parameters are given for comparison in table A.1.

The fits are about equally good. However, on physical grounds the sinusoidal model with a significant c-axis component on the cubic sites is difficult to understand. How can a magnetic order in the hexagonal planes induce a perpendicular order on the cubic sites? The susceptibility measurements [23] show there is a planar anisotropy on the cubic sites. This would not favour a moment component along the c-axis. The present spiral model avoids these problems, and is hence physically more acceptable. It may be a $1q$ or a multi-q structure; the available data cannot tell.

The remarkable observation by Watson *et al* [11] was that only two pairs of satellites were observed by x-rays out of the possible six. This proved definitely that Nd does not have triple-q structure at $T = 11.5$ K in the probed region.

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