

For the commensurate magnetic phase of  $\text{NdFe}_3(^{11}\text{BO}_3)_4$  we measured the polarization matrix at six magnetic Bragg reflections and at  $T = 20, 25$  and  $30$  K. The measured polarization matrices proved to be independent of temperature (within the error bars) for  $T \geq 20$  K. Thus, only the data for  $T = 20$  K will be discussed in the following. The corresponding matrices are provided in table II. From the measured polarization matrices several constraints on the magnetic structure can be derived:

1. On all measured Bragg peaks the elements  $yx$  and  $zx$  are equal to zero. This implies that the magnetic structure is not chiral at all or that it is a chiral structure with equally populated chiral domains.
2. For the magnetic reflection  $(0,0,-1.5)$  the scattering vector  $\mathbf{Q}$  is directed parallel to the crystallographic  $c$  direction. Hence, the magnetic interaction vector only contains components in the basal plane.  $\mathbf{P}'_{xx} = -|\mathbf{M}_\perp|^2/|\mathbf{M}_\perp|^2 = -0.872(2)$ . Here the reduction from  $-1$  is due the polarizing benders which have a non-ideal polarization efficiency of approximately  $0.966$ . Thus,  $\mathbf{P}'_{xx}$  is fully polarized whereas  $\mathbf{P}'_{yy} = (|\mathbf{M}_{\perp y}|^2 - |\mathbf{M}_{\perp z}|^2)/|\mathbf{M}_\perp|^2$  and  $\mathbf{P}'_{zz} = (-|\mathbf{M}_{\perp y}|^2 + |\mathbf{M}_{\perp z}|^2)/|\mathbf{M}_\perp|^2$  are fully depolarized. This would foremost lead to the assumption  $|\mathbf{M}_{\perp y}|^2 \approx |\mathbf{M}_{\perp z}|^2$  but as the elements  $\mathbf{P}'_{yz}$  and  $\mathbf{P}'_{zy}$  are also equal to zero it also suggests the presence of spin domains in the basal plane.
3. On the magnetic reflection  $(0,4,0.5)$  the scattering vector  $\mathbf{Q}$  is approximately parallel to the reciprocal  $b^*$  axis. As the  $z$  axis which is perpendicular to the scattering plane lies always within the basal hexagonal plane in the chosen scattering geometry the  $y$ -axis is approximately parallel to the crystallographic  $c$ -axis ( $\angle(y, c) \approx 15^\circ$ ). The polarization tensor shows  $\mathbf{P}'_{xx} \approx \mathbf{P}'_{yy} \approx -\mathbf{P}'_{zz}$  which indicates that  $|\mathbf{M}_{\perp y}|^2 \approx 0$  and hence the magnetic interaction vector is directed along  $z$ . Therefore the magnetic moments are confined in the basal plane.

We note that these constraints are satisfied by both possible magnetic structures (M1a/b) and (M2). In order to calculate the expected polarization matrices for both models, we used lattice constants and structural parameters from table I. The magnitude of the magnetic moments for the Fe and Nd ions were set to the values as obtained from the unpolarized diffraction data. Since SNP is generally insensitive to absolute moment sizes in case of pure magnetic reflections they were fixed in subsequent fits. Both models fail to explain the observed polarization matrices when no orientation domains were considered. Introducing the three or six orientation domains with statistical population in the calculation for models (M1) and (M2), respectively (cf. appendix), resulted in polarization matrices given in table II. Further, we considered the non-ideal polarization efficiency of the used polarizing benders in the

calculation.

As demonstrated in table II the magnetic model (M2) was not able to explain the measured polarization tensors on the two magnetic reflections  $(0, 1, -2.5)$  and  $(0, 1, 0.5)$  as indicated by the bold entries in table II ( $\chi^2 = 9.5$ ). The model introduces small chiral contributions ( $yx$  and  $zx$  elements of the tensors) due to its slightly canted spins (cf Fig. 1(b)), that are not observed in the experiment. Performing a fit from this starting values did not result in a better agreement between model and data, as the fit diverged. Hence, model (M2) can be excluded.

For the magnetic model (M1) we fixed  $\phi_{\text{Fe}} = 0$  and left  $\phi_{\text{Nd}}$  free for the fits, thus corresponding to (M1b). The fit converges to a solution with  $\phi_{\text{Nd}} = 0$  ( $\chi^2 = 3.8$ ) which corresponds to (M1a). Attempts to additionally determine the orientation of the Fe magnetic moments via the angle  $\phi_{\text{Fe}}$  gave no conclusive results. The angle  $\phi_{\text{Fe}}$  describes the absolute orientation of the magnetic moments in the crystallographic  $ab$ -plane with respect to chemical structure. Thus, the indeterminacy of  $\phi_{\text{Fe}}$  is most probably related to the presence of the three orientation domains in the hexagonal basal plane.

In summary our SNP data for the commensurate phase is best explained by the model (M1a) when  $\phi_{\text{Fe}} = \phi_{\text{Nd}} = 0$  which is also in agreement with the unpolarized neutron single crystal diffraction results described in section III C. The corresponding magnetic structure is illustrated in Fig. 1(a).

TABLE III: (a) Integrated intensities for the magnetic satellite reflections in the IC phase as calculated from model (M1a), however, with the incommensurate  $\mathbf{k}_{hex,i}$ . For the calculation we assumed that only one single chiral domain is populated. (b) Measured integrated intensities for the  $\mathbf{Q}$ -scan in Fig. 5(a) in the different polarization channels. For the comparison see the text.

(a)				
Peak	$I_{xx}^c$	$I_{x-x}^c$	$I_{-xx}^c$	$I_{-x-x}^c$
$(0,0,0)-\mathbf{k}_{hex,i}$	4.15	0.17	99.56	4.15
$(0,0,-3)+\mathbf{k}_{hex,i}$	4.16	99.83	0.17	4.16
(b)				
Peak	$I_{xx}^m$	$I_{x-x}^m$	$I_{-xx}^m$	$I_{-x-x}^m$
integrated over $\pm\mathbf{k}_{hex,i}$	66(8)	934(31)	946(31)	65(8)

## B. Chirality in the incommensurate magnetic phase

Fig. 5(a) shows polarized constant-energy-scans along the  $L$ -direction performed around the reciprocal space position  $(0, 0, -1.5)$  at  $T = 1.5$  K in the IC-phase. Four different polarization channels were measured, namely  $I_{xx}$ ,  $I_{x-x}$ ,  $I_{-xx}$  and  $I_{-x-x}$ . The scans for the chan-