Preferred mineral orientation of a chloritoid-bearing slate in relation to its magnetic fabric

Tom Haerinck^{a,*}, Hans-Rudolf Wenk^b, Timothy N. Debacker^c, Manuel Sintubin^a

a. Department of Earth & Environmental Sciences, KULeuven, Celestijnenlaan 200E, B-3001 Leuven, Belgium

b. Department of Earth and Planetary Science, University of California, Berkeley, CA 94720, U.S.A.

c. FROGTECH Ltd., 2 King Street, 2600 Deakin West, ACT, Australia

** Corresponding author: tom.haerinck@ees.kuleuven.be; tel. +32 16 372 191*

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Abstract

A regional analysis of the anisotropy of the magnetic susceptibility on low-grade metamorphic, chloritoid-bearing slates of the Paleozoic in Central Armorica (Brittany, France) revealed very high values for the degree of anisotropy (up to 1.43). Nonetheless, high-field torque magnetometry indicates that the magnetic fabric is dominantly paramagnetic. Chloritoid's intrinsic degree of anisotropy of 1.47 ± 0.06 , suggests that chloritoid-bearing slates can have a high degree of anisotropy without the need of invoking a significant contribution of strongly anisotropic ferromagnetic (s.l.) minerals. To validate this assumption we performed a texture analysis on a representative sample of the chloritoid-bearing slates using hard X-ray synchrotron diffraction. The preferred orientation patterns of both muscovite and chloritoid are extremely strong (~ 38.6 m.r.d. for muscovite, 20.9 m.r.d. for chloritoid) and display roughly axial symmetry about the minimum magnetic susceptibility axis, indeed suggesting that chloritoid may have a profound impact on the magnetic fabric of chloritoid-bearing rocks. A calculation of the anisotropy of the magnetic susceptibility tensor of a 3-phase model of the slate, based on the mineral textures and their magnetic single crystal tensors reveals a

significant discrepancy with the measured magnetic anisotropy of the investigated slate.

1. Introduction

The anisotropy of magnetic susceptibility (AMS) of a rock depends on the orientation distribution of the rock-forming minerals and their intrinsic magnetic properties (shape or magnetocrystalline anisotropy). Commonly, several magnetic carriers, with ferromagnetic (*sensu lato* - *s.l.*, i.e. a combining term for ferromagnetic *sensu stricto*, ferrimagnetic and antiferromagnetic), paramagnetic and/or diamagnetic behavior, contribute to the magnetic fabric. The diamagnetic fabric will only be important when the other contributions are very weak, such as in the case of pure sandstones and limestones (Borradaile et al., 1999; de Wall et al., 2000). While the ferromagnetic (*s.l.*) contribution to the magnetic fabric primarily depends on domain state, grain size and interaction between particles, the paramagnetic contribution results from intrinsic crystal lattice anisotropy of the paramagnetic minerals and their degree of preferred orientation (Borradaile and Jackson, 2010). Therefore, the paramagnetic fabric is more likely to serve as a proxy for the petrofabric related to the deformation of the rock in slates.

Although many pioneering magnetic fabric studies assumed ferromagnetic (*s.l.*) minerals to be the primary cause for the observed magnetic fabrics in fine-grained siliciclastic metasedimentary rocks (Graham, 1954; Rees, 1961; Fuller, 1964), Fe-bearing phyllosilicates often are the main magnetic carriers contributing to the magnetic fabric (e.g. Coward and Whalley, 1979; Borradaile et al., 1986; Rochette, 1987). For Fe-bearing phyllosilicates, it was found that the degree of intrinsic magnetocrystalline anisotropy (P_J) does not exceed 1.35 (Beausoleil et al., 1983; Borradaile et al., 1987; Zapletal, 1990; Borradaile and Werner, 1994; Martín-Hernández and Hirt, 2003) (see Table 1 for definition of the AMS parameters used in this work). Consequently, the P_J value of 1.35 has been used as the upper limit for the paramagnetic contribution to the AMS of siliciclastic metasedimentary rocks and higher P_J values are systematically attributed to a ferromagnetic (*s.l.*) contribution (Rochette, 1987; Rochette et al., 1992).

A regional magnetic fabric study of chloritoid-bearing slates of the Palaeozoic Plougastel formation in the low-grade metamorphic (epizone) Monts d'Arrée slate belt (MASB) in Central Armorica (Brittany, France) (Figure 1), revealed a dominantly paramagnetic fabric with P_J values up to 1.43. In contrast, the stratigraphically equivalent slates free of chloritoid, in the very low-grade (anchizone) Crozon fold-

and-thrust belt, showed P_J values only up to 1.27 (Haerinck et al., 2013a). In order to assess the possible paramagnetic contribution of monoclinic chloritoid, its magnetocrystalline anisotropy has been determined on a collection of chloritoid crystals, collected from different tectonometamorphic settings worldwide (Haerinck et al., 2013b). The magnetocrystalline degree of anisotropy of chloritoid was found to be 1.47 ± 0.06 , which is significantly higher than the magnetocrystalline degree of anisotropy of most paramagnetic phyllosilicates (1.35, references above). Furthermore, a new analysis of the magnetocrystalline anisotropy of muscovite single crystals by Biedermann *et al.* (2014) revealed a P_J value of 1.44 ± 0.02 , much higher than previously supposed (1.15 ± 0.05 ; Martín-Hernández and Hirt, 2003).

These very high magnetocrystalline degrees of anisotropy suggest that the very strong paramagnetic anisotropy of the chloritoid-bearing slates of the MASB may be due to a (very) strong alignment of the chloritoid and potentially also muscovite crystals. To our knowledge, there are no examples of a quantitative texture analysis on chloritoid-bearing slates, making it impossible to check the validity of our basic assumption. Therefore, the presumed strong chloritoid alignment in the slates of the MASB was the incentive to study the preferred orientation of the rock-forming minerals in a representative sample of these slates.

The preferred orientation of phyllosilicates in argillaceous metasedimentary rocks has been studied primarily by X-ray pole figure goniometry (e.g. Wood and Oertel, 1980; Oertel, 1983; Sintubin, 1993; van der Pluijm et al., 1994). Since then, new methods have been developed to obtain quantitative information from diffraction images produced by high energy synchrotron X-rays (e.g. Wenk et al., 2010). A major advantage of the synchrotron X-ray method is that information is obtained about the full crystal preferred orientation (CPO) of all minerals composing a bulk sample. Here we present the preferred orientation of the constituting minerals muscovite, chloritoid, chlorite and quartz of a representative sample of the chloritoid-bearing slate of the MASB to explore whether the pronounced magnetic anisotropy in this chloritoid-bearing slate can indeed be attributed to a strong alignment of chloritoid crystals. Note that such a link between mineral preferred orientations and magnetic properties is well established for slates in which the magnetic properties are dominated by the paramagnetic phyllosilicate minerals (e.g. Richter et al., 1993; Siegesmund et al., 1995; Chadima et al., 2004; Hansen et al., 2004; Martín-Hernández et al., 2005; Cifelli et al., 2009; Oliva-Urcia et al., 2010).

2. Sample Characterization and Magnetic Properties

We investigated a representative sample of chloritoid-bearing slates typical for the low-grade (epizone) metamorphic MASB, a particular Variscan tectonostratigraphic segment of the Central Armorica Domain (CAD) in Brittany (France) (Figure 1). The CAD represents a part of the Perigondwanan microcontinent Armorica and consists of a Neoproterozoic, Cadomian cratonic basement, reflecting Pan-African geodynamics (Ballèvre et al., 2001; Chantaine et al., 2001), and its late Proterozoic to Palaeozoic metasedimentary cover (Guillocheau and Rolet, 1982; Guerrot et al., 1992). The MASB is primarily composed of rocks of the Pridolian to Lochkovian Plougastel Formation and is characterized by a high-strain deformation that occurred during a single, progressive, NW-SE oriented, contractional deformation event in low-grade metamorphic conditions (Sintubin et al., 2008). This coaxial, homogeneous shortening is accommodated by both cylindrical folding and cogenetic cleavage development, and is estimated to be in the order of 50 to 60 %, which reflects the bulk regional strain (van Noorden et al., 2007). The sample location (coordinates: N48° 24' 22.970" – W03° 54' 42.579") is an outcrop with a NW-SE oriented outcrop face of approximately 20 m long and 10 m high, located 100 m west of Roc'h Trévél (Figures 1-2). The outcrop displays a multimetre-scale antiform and consists of a multilayer sequence with relatively thin quartzitic and pelitic layers, a few competent quartzitic sandstone beds and two homogeneous siltstone beds (HSB). The sample (sample BR09TH131C3) has been taken from the HSB in the core of the antiform (Figure 2a-b).

The HSBs of the Roc'h Trévél outcrop area show a strongly developed cleavage fabric and lack any macroscopically visible bedding fabric or grain-size variation. An intersection lineation is often visible on the cleavage planes, having a subhorizontal NE-SW trending orientation. Microscopic fabric analysis shows that the strongly developed tectonic cleavage is a spaced foliation, consisting of cleavage domains with micaceous material and chloritoid minerals and microlithons containing primarily quartz (Figure 2c). Mineralogical analysis of the HSBs of the Roc'h Trévél outcrop area shows that they are dominated by quartz (42 ± 9 vol%) and white mica (37 ± 10 vol%), with a significant fraction of chloritoid (18 ± 5 vol%) and a minor amount of chlorite (2 ± 2 vol%) (Haerinck et al., 2013a).

The low-field and high-field AMS is measured on a cubic sample of 8 cm³ cut from an in situ oriented homogeneous siltstone sample with one face parallel to the cleavage plane (Figure 2b insert). It is

defined by a Cartesian sample coordinate system with A along the cleavage's strike, B along the cleavage's dip and C along the pole to the cleavage. The low-field AMS analysis, using an AGICO KLYS3 kappabridge (Jelínek and Pokorný, 1997) at the KU Leuven, shows a bulk magnetic susceptibility (K_m) of 389×10^{-6} [SI]. The magnetic susceptibility ellipsoid is strongly oblate, as evidenced by a shape parameter (T) of 0.76, and has an extremely strong eccentricity, evidenced by a corrected degree of anisotropy (P_J) of 1.40. The minimum magnetic susceptibility axis (K_3) shows a close angular relationship with both the pole to bedding and the pole to cleavage (sample's C axis), (Figure 3): both poles are oriented at an angle of 6° with respect to K_3 , which is roughly equal to the accuracy of the sampling and preparation procedure. However, a regional analysis of the direction of K_3 with respect to the macroscopic fabric elements in the MASB (e.g. Haerinck et al., 2013a) shows that the triaxial to oblate magnetic fabrics are consistently parallel to the tectonic cleavage, and not to the bedding fabric. The maximum magnetic susceptibility axis (K_1) is subhorizontal and NE-SW directed, and approximates both the sample's A axis (difference of 3°) and the fold hinge lines and bedding-cleavage intersection lineation in the outcrop (Figure 3). Note that the calculated bedding-cleavage intersection of the sample plunges to the south, and thus is oblique with respect to K_1 (Figure 3). However, the small angle between bedding and cleavage in combination with the poorly constrained bedding orientation, makes this calculated intersection orientation very uncertain. The intermediate susceptibility axis (K_2) coincides more or less with the dip direction of the cleavage, i.e. the sample's B axis (difference of 5°).

The sample's high-field AMS was measured at ETH Zürich with a torsion magnetometer (Bergmüller et al., 1994) in fields from 1000 to 1500 mT (Figure 4), and processed using the mathematical method of Martín-Hernández and Hirt (2001, 2004), which allows a separation of the paramagnetic and ferromagnetic (s.l.) contribution to the sample's magnetic anisotropy. The direction of the principal susceptibilities of the paramagnetic component ($K_{1,2,3}^{hf-p}$) corresponds closely to those of the low-field AMS, i.e. the difference is 9° , 12° and 10° , respectively (Figure 3). On the other hand, the principal susceptibilities of the ferromagnetic (s.l.) component ($K_{1,2,3}^{hf-f}$) are markedly oblique to those of the low-field AMS and seemingly unrelated to the macroscopic fabric elements (Figure 3). A calculation of the contribution of both components to the sample's magnetic anisotropy clearly shows that the paramagnetic component dominates ($88 \pm 6\%$) over the ferromagnetic (s.l.) component ($12 \pm 3\%$). Because the torque magnetometry approach can only calculate the deviatoric part of the susceptibility

tensor of both components, i.e. the difference of the AMS ellipsoid from a sphere, an independent determination of the paramagnetic bulk susceptibility is required to calculate the full tensor of the isolated paramagnetic component and its anisotropy parameters ($P_J^{\text{hf-p}}$ and $T^{\text{hf-p}}$).

Unfortunately we do not have this information for sample BR09TH131C3. Instead, we can use the bulk susceptibility of the low-field AMS (K_m) analysis to calculate an absolute minimum value for $P_J^{\text{hf-p}}$ and $T^{\text{hf-p}}$. This way we actually suppose that the paramagnetic susceptibility is equal to K_m . If there really is a ferromagnetic (s.l.) contribution to K_m , the paramagnetic susceptibility will be lower than K_m , and $P_J^{\text{hf-p}}$ and $T^{\text{hf-p}}$ will increase. By using this approach, we obtain a minimum value of 1.37 for $P_J^{\text{hf-p}}$ and 0.80 for $T^{\text{hf-p}}$, respectively. Alternatively, we can calculate a maximum theoretical paramagnetic susceptibility (MTPS) from the Fe^{2+} , Fe^{3+} and Mn^{2+} composition of the sample (Haerinck et al., 2013a and references therein). Because the chemical analysis is performed on only 10 mg of the crushed sample, and we do not know how representative this is for the entire sample of 25 g, the obtained MTPS values can only serve as an estimate for the paramagnetic susceptibility. The analysis was performed twice, on different powder samples collected from the crushed sample, resulting in a MTPS of 280×10^{-6} [SI] (3.85 wt% FeO, 0.22 wt% Fe_2O_3 and 0.03 wt% MnO) and 311×10^{-6} [SI] (4.33 wt% FeO, 0.19 wt% Fe_2O_3 and 0.03 wt% MnO), respectively. These values are significantly lower than K_m (389×10^{-6} [SI]), and consequently result in higher $P_J^{\text{hf-p}}$ values of 1.56 and 1.49, respectively. So, this paramagnetic, high-field degree of anisotropy is definitely above the 'classical' upper limit of 1.35 for paramagnetic phyllosilicates (Rochette, 1987; Rochette et al., 1992).

3. Preferred Orientation Analysis

The hard X-ray synchrotron diffraction measurements were done at the high-energy beamline BESSRC 11-ID-C of APS (Advanced Photon Source) of Argonne National Laboratory. A monochromatic X-ray beam with a wavelength of 0.10798 Å and a beam diameter of 1 mm was used. Diffraction images were recorded with a Perkin Elmer amorphous silicon detector (2048 x 2048 pixels) at a distance of about 200 cm from the sample. The sample is a 1 mm thick slab cut perpendicular to the cleavage plane, containing both the sample's B and C axis (Figure 2b insert), and mounted on a goniometer with an axis perpendicular to the incident beam. The incident beam (at 0° tilt) parallels the sample's A axis. The slab was rotated in 15° increments around the sample's C axis, from -45° to

+45°. At each rotation position a diffraction image was recorded while the sample was translated 2 mm parallel to the rotation axis (~C) to obtain a representative volume average. The rotations are necessary to obtain adequate pole figure coverage for determining preferred orientation. Figure 5 shows the diffraction image at 0° tilt and axes C and B are indicated. Intensity variations along Debye rings are indicative of preferred orientation which is strong for phyllosilicates, e.g. muscovite M002 and chloritoid Ct004 and weak for quartz Q100.

For an estimation of the mineralogical composition and preferred orientations we relied on the Rietveld refinement of diffraction images, implemented in the software MAUD (Lutterotti et al., 1997; Lutterotti et al., 2014). The Rietveld method obtains a best fit between measured diffraction patterns and a model based on a number of refined parameters, such as instrument geometry, phase volume fractions, unit cell parameters, preferred orientations, etc. Seven diffraction images, measured at different sample tilts (-45°, -30°, -15°, 0°, 15°, 30°, 45°), were used and integrated over 10° sectors to obtain diffraction patterns. For the Rietveld analysis a scattering angle 2θ range from 0.3° to 3.5° was applied. Four mineral phases were considered, with corresponding crystallographic information from the literature: monoclinic muscovite (Guggenheim et al., 1987; amcsd #0001076), monoclinic (Koch-Müller et al., 2000; amcsd #0006829) and triclinic chloritoid (Hanscom, 1980; amcsd #0000786), monoclinic chlorite (Zanazzi et al., 2007; amcsd #0004284) and trigonal quartz (Antao et al., 2008; amcsd #0006212). Preferred orientation was refined with the E-WIMV algorithm and an orientation distribution cell size of 10°. Orientation distributions were exported from MAUD and further processed in BEARTEX (Wenk et al., 1998) to calculate and plot pole figures. For the Rietveld texture analysis of the monoclinic phyllosilicates the first setting (c-axis rotation axis) has to be used (Matthies and Wenk, 2009), but for all labels of Miller indices in pole figures, we use the more common second setting (b-axis rotation axis and (001) as cleavage plane).

Figure 6 shows two diffraction patterns, one parallel and the other perpendicular to the foliation, i.e. along the sample's B and C axis, respectively. They show very different intensities. The pattern perpendicular to the foliation (top) shows very high intensities for basal plane reflections of the phyllosilicates (M002, Ch002, M004, Ct004). There is excellent agreement between the measured spectrum (crosses on Figure 6) and the Rietveld model (solid line on Figure 6). This is even more obvious in Figure 7, presenting a stack of all diffraction patterns for an image with experimental data at

the bottom and the Rietveld fit on top. Debye rings in the image in Figure 5 are here expressed as straight lines.

The Rietveld refinement provides volume fractions of the different phases (Table 2). The slate is dominated by quartz (45 vol%) and muscovite (42 vol%), with a significant fraction of chloritoid (13 vol%) and only a small fraction of chlorite (1 vol%). These values conform very well with the composition determined by Haerinck *et al.* (2013a) for other homogeneous siltstone samples collected in the Roc'h Trévezel site.

Pole figures in equal area projection on the foliation plane of the different phases are shown in Figures 8 and 9. Contours express pole densities in multiples of a random distribution (m.r.d.). The pole figures for muscovite and chloritoid display very strong maxima for (001) perpendicular to the foliation, indicative of a strong shape preferred orientation of both muscovite (38.6 m.r.d.) and chloritoid (20.9 m.r.d.) within the slaty cleavage plane. The orientation distribution patterns are basically axially symmetric with no significant alignment of poles such as (100) or (010) in the cleavage plane. Small deviations from axial symmetry are attributed to incomplete pole figure coverage. Although much weaker, chlorite shows a similar orientation distribution (5.8 m.r.d.). The preferred orientation patterns of quartz are weak (< 1.5 m.r.d.). The most striking features, though, are a maximum of positive rhombs (101) perpendicular to the foliation (1.55 m.r.d.) and a concentration of quartz a-axes (110) in the plane of foliation and parallel to the lineation. Furthermore, the quartz c-axes (001) form a complex small circle girdle with a distinct minimum perpendicular to the foliation.

4. Discussion

With synchrotron X-ray diffraction preferred orientation distributions of complex polymineralic rocks such as slates can be quantified, which is not possible with traditional pole figure goniometry. By analyzing diffraction images with the Rietveld method we can obtain separate pole figures for any crystallographic direction for muscovite, chloritoid, chlorite and quartz, as well as volume fractions of the phases. This provides a basis for comparing microstructural fabric characteristics with bulk magnetic properties.

The axial pole figure patterns of muscovite, chlorite and chloritoid (Figure 8) can be interpreted to reflect a pure flattening strain related to the slaty cleavage development. These pole figure patterns do

not bear any evidence for the development of a stretching lineation, nor is there any indication of an intersection lineation resulting from two different orientation populations (cf. Sintubin, 1996; Sintubin, 1998), regardless of the macroscopic presence of a bedding/cleavage intersection lineation. It is remarkable that the preferred orientations of both muscovite (38.9 m.r.d.) and chloritoid (20.9 m.r.d.) is extremely high with respect to other slates, such as very low-grade to low-grade metamorphic shales and slates in the Palaeozoic of the Brabant-Ardenne area (Belgium, France; Sintubin, 1994; Debacker and Sintubin, 2008) and in the high-strain Cambrian slate belt of Wales (Wood and Oertel, 1980).

Experimental studies of fabric formation in fine-grained sediments have shown that mechanical rotation of pre-existing grains is only capable of producing fabrics up to about 10 m.r.d. (Tullis, 1976; Haines et al., 2009). Hence, the extremely strong muscovite and chloritoid texture in the sample of the MASB has to result from a preferred metamorphic mineral growth in response to a differential stress (cf. van der Pluijm et al., 1998; Ho et al., 2001). This is conform with the syntectonic character of the chloritoid growth that has been observed throughout the MASB (Darboux and Garreau, 1976; Darboux, 1991). Syntectonic growth, related to a pure flattening tectonic strain, may also explain the axial symmetry of the pole figure patterns of both phyllosilicates (muscovite, chlorite) and chloritoid.

With respect to the quartz pole figure patterns (Figure 9), it is striking that a maximum of the positive rhombs 101 is oriented perpendicular to the slaty cleavage, i.e. parallel to the overall shortening direction. This could be due to mechanical Dauphiné twinning which aligns the direction of greatest elastic compliance, orthogonal to the positive rhombs, with the maximum compressive principal stress direction (e.g. Tullis, 1970), but this would not explain the quartz c-axis preferred orientation. Alternatively, it could be caused by dynamic recrystallization under stress with growth of crystals in thermodynamically favorable directions (e.g. Kamb, 1959). Crystal-plastic deformation features in quartz grains (irregular shape, lobate boundaries, undulose extinction) indicate that recrystallisation of quartz grains is also syntectonic.

In order to investigate the relationship between texture and magnetic anisotropy of the slate, we compared (1) the orientation of the magnetic fabric to the mineral's CPO and (2) the anisotropy of the measured AMS tensors of the slate to that of the calculated magnetic susceptibility tensor of a 3-phase model of the slate. The latter is obtained, firstly, by calculating the magnetic susceptibility tensors for the muscovite, chloritoid and chlorite phase based on the CPOs and single crystal tensors, and secondly, by taking their weighted average according to the relative volume percentage of the

different phases, i.e. 42 % muscovite, 13 % chloritoid and 1 % chlorite. Quartz has a magnetic susceptibility of -14×10^{-6} [SI] (Hrouda and Kapička, 1986). Therefore, the quartz contribution (41 %) does reduce the overall strength of the calculated magnetic tensor but has no significant influence on its anisotropy. The magnetic 2nd rank tensor calculations are done using BEARTEX (Wenk *et al.*, 1998). The single crystal tensor of chloritoid is derived from single crystal measurements of Haerinck *et al.* (2013b), those from muscovite and chlorite from single crystal measurements of Biedermann *et al.* (2014) (Table 3).

Both the orientation of the minimum magnetic susceptibility axis, K_3 and K_3^{hf-p} , and the preferred orientation maximum of (001) for muscovite, chloritoid and chlorite are sub-parallel to the sample's C axis, hence parallel to the cleavage pole (Figure 2b). As the CPOs of muscovite, chloritoid and chlorite are axially symmetric and the magnetic susceptibility ellipsoid is strongly oblate, it follows that the mineral and magnetic fabric are sub-parallel. The magnetic data also show a weak lineation, i.e. there is a clearly defined K_1 (and K_1^{hf-p}) that is oriented within the cleavage plane and coincides roughly with the orientation of the macroscopically observed bedding/cleavage intersection lineation (i.e. sample's A axis). This deviation from the (nearly) perfect oblate nature of the magnetic fabric in contrast to the axially symmetric mineral textures could be caused by the monoclinic crystal structures of muscovite, chloritoid and chlorite. The crystallographic b-axis of these monoclinic crystals (second setting) possesses the two-fold symmetry and thus has to be parallel to one principal axis of the susceptibility ellipsoid (Borradaile and Jackson, 2010). Hence, the crystal's a- and c-axis are perpendicular to the b-axis and inclined with respect to the other two (orthorhombic) AMS axes. Indeed, Martín-Hernández (2002) observed an angle of $3^\circ \pm 3^\circ$ between K_3 and the pole of the basal plane for both muscovite and chlorite, and Haerinck *et al.* (2013b) show that there is an angle of $5^\circ \pm 2^\circ$ between K_3 and the pole of the chloritoid basal plane. Consequently, even though the constituent paramagnetic minerals, muscovite, chlorite and chloritoid, have a nearly perfectly axially symmetrical CPO, there will be a slight dispersion of the minimum susceptibility axes, resulting in the small deviation between the magnetic and sample axes and a less oblate shape of the slate's magnetic fabric with respect to that of the mineral constituents: T of 0.76 and T^{hf-p} of 0.80 for the sample's susceptibility ellipsoid vs. 0.84, 0.95 and 0.94 for the muscovite, chlorite and chloritoid ellipsoids, respectively (Martín-Hernández and Hirt, 2003; Haerinck *et al.*, 2013b).

For the calculated 3-phase model, we obtain a bulk susceptibility that is a bit lower than the measured value (Table 3). This may be due to the variability of the bulk susceptibility of the single crystals, depending on the crystals' cation content (Haerinck et al., 2013b; Biedermann et al., 2014). The shape parameter T of the calculated 3-phase model is very similar to the measured value. On the other hand, the corrected degree of anisotropy (P_J) of the calculated 3-phase model is clearly lower than the measured values, i.e. 1.21 instead of 1.40 and 1.37 for the low-field and high-field AMS, respectively (Table 3). The calculated tensor is strongly dominated by the chloritoid phase, because of its high bulk susceptibility compared to muscovite and chlorite. The anisotropy of this chloritoid phase (1.20) is, though, significantly reduced with respect to the anisotropy of chloritoid single crystals (1.47) because the chloritoid texture shows a relatively high background of randomly oriented crystallites (0.5 m.r.d., Table 2), indicating that chloritoid occurs more randomly than muscovite. Therefore, the orientation distribution of chloritoid considerably deviates from a 'near perfect' preferred orientation mimicking a single crystal.

A similar problem has been faced by Biedermann (2014), who calculated the magnetic properties of metamorphic rocks from amphibole and pyroxene texture data. They found that the recovered magnetic anisotropy in terms of k' (Jelinek, 1984) often represents only 40 to 60 % of the measured anisotropy. This is similar to our recovered magnetic anisotropy in terms of k' , which is 40 and 44 % of the measured low-field and high-field value, respectively. For now, the authors have to conclude that based on the comparison of calculated and observed magnetic anisotropy, the weighted average of the magnetocrystalline anisotropy of the chloritoid, muscovite and chlorite phases seems not sufficient to explain the measured magnetic anisotropy. A number of possible explanations can be considered:

- 1) There is another phase contributing to the magnetic anisotropy and the CPO of this phase is not detected and thus not taken into account when calculating the 3-phase model. This potential phase must be present in a very low quantity but have a profound impact on the slate's magnetic anisotropy. Moreover, its contribution to the magnetic anisotropy would not be separated from the paramagnetic contribution by the high-field method, implying that no saturation occurs in the field range used. Secondly, its contribution to the bulk susceptibility must be rather low. An antiferromagnetic phase, e.g. goethite or hematite, could meet these criteria.

- 2) The muscovite phase is relatively Fe-rich (e.g. phengite) and consequently has a higher intrinsic magnetic susceptibility than the muscovite single crystal tensor of Biedermann *et al.* (2014), used in the current calculations. Compared to chloritoid (see above), the anisotropy of the muscovite phase (1.33) is relatively less reduced with respect to the anisotropy of muscovite single crystals (1.44), which may be explained by the 'near perfect' preferred orientation of muscovite, mimicking a single crystal (Figure 8). Therefore, a higher intrinsic bulk magnetic susceptibility of the muscovite crystals would result in an increase of the slate's calculated magnetic anisotropy.
- 3) The chloritoid single crystal tensor of Haerinck *et al.* (2013b), used in the current calculation, underestimates the degree of anisotropy of the chloritoid crystals in the slates of the MASB. Taking into account the ratio of the calculated chloritoid phase P_J to the currently used chloritoid single crystal P_J (Table 3), we would actually need an chloritoid single crystal P_J of 1.66 to obtain a good match between the measured magnetic anisotropy and the anisotropy of the 3-phase model. Note that the chloritoid single crystal tensor of Haerinck *et al.* (2013b) is an average of 7 different chloritoid crystals. These crystals do show some variation in P_J . However, the highest P_J measured for a chloritoid single crystal was 1.54, which is still significantly lower than the required P_J of 1.66. Possibly, the obtained magnetic anisotropy for chloritoid single crystals by Haerinck *et al.* (2013b) underestimates the magnetocrystalline anisotropy of chloritoid if the analyzed single crystals (from 7 different tectonometamorphic settings) are in fact no single crystals, but are systematically twinned.
- 4) There is another factor contributing to the measured magnetic anisotropy apart from the magnetocrystalline anisotropy of different (para)magnetic phases. This additional component of the observed magnetic anisotropy could be a distribution anisotropy. This phenomenon occurs if anisotropic magnetostatic interactions among grains affect the bulk sample properties (Borradaile and Jackson, 2010). In literature, it is exclusively associated with ferromagnetic (*s.l.*) particles (e.g. Hargraves et al., 1991; Stephenson, 1994). However, as the intrinsic bulk susceptibility of the chloritoid crystals is relatively high ($K_m = 1726 \times 10^{-6}$ [SI]) and the chloritoid crystals are closely spaced in the cleavage domains (Figure 2c), a relevant magnetostatic interaction might be produced between the chloritoid crystals in the slate's cleavage domains.

5. Conclusion

In the investigated sample of the chloritoid-bearing slates of the Monts d'Arrée slate belt in Brittany, both muscovite and chloritoid display a very strong preferred orientation of the basal planes parallel to the macroscopic cleavage fabric. The axes of the crystal orientation pattern are parallel to the measured magnetic fabric, both the one obtained using low-field AMS and the isolated paramagnetic fabric derived from the high-field approach. At first, comparing the magnetic fabric and the mineral textures clearly illustrates that due to its relatively high magnetic susceptibility and very strong magnetocrystalline anisotropy ($P_J = 1.47$), chloritoid may have a profound impact on the magnetic fabric of chloritoid-bearing rocks.

On the other hand, a calculation of the anisotropy of the magnetic susceptibility tensor of a 3-phase model of the slate shows that the weighted average of the magnetocrystalline anisotropy of the chloritoid, muscovite and chlorite phases seems not sufficient to explain the measured magnetic anisotropy of the investigated slate. The reason for this discrepancy still remains unclear to date, although a number of possible explanations are considered. To further establish this relationship similar studies on chloritoid-bearing slates and schists should be performed in the future.

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Figure captions

Figure 1 – Location of the Roc'hTrévél outcrop area in the MASB in Central Armorica, Brittany, France (modified after Chantraine et al., 1981; Hallégouët et al., 1982; Castaing et al., 1987).

Figure 2 – Outcrop MA006 (coordinates: N48° 24' 22.970" – W03° 54' 42.579") (see Figure 1 for location). **(a)** photograph of the outcrop. **(b)** Line drawing with indication of the homogeneous siltstone beds and sample locations. The insert shows the sample BR09TH131 and the cubic subsample C3 on which the magnetic measurements have been performed. The Cartesian sample coordinate systems (A, B, C) and the slab cut for the synchrotron diffraction measurement (grey) are indicated. **(c)** Micrograph of sample BR09TH133 (position in Figure 2b) showing a spaced foliation with cleavage domains consisting of micaceous material.

Figure 3 – Magnetic anisotropy parameters of sample BR09TH131C3. Abbreviations: EV – eigenvalues of the AMS tensor; Dir'n: direction of the AMS eigenvalues or macroscopic fabric element (trend / plunge or dip direction / dip).

Figure 4 – Results of the high-field torque analysis of specimen BR09TH131C3. Rotation positions 1 and 2 (around the sample's B and A axis) show a well-defined periodical function with maximum and minimum values at approximately 45°, 135°, 225° and 315°, i.e., when the angle between the induced field and the cleavage plane is 45°. Rotation position 3 (around the sample's C axis) shows a very weak torque signal. The plot on the right shows that the amplitude of the periodical torque function increases linearly as a function of the inducing magnetic field squared (B^2) for the three measurement positions. The intercept close to zero indicates a dominance of the paramagnetic contribution to the high-field AMS, i.e. 88 ± 6 %. The calculated deviatoric eigenvalues, i.e. the absolute difference between the mean susceptibility and the principal susceptibilities, are given for both the paramagnetic and ferromagnetic (s.l.) component to the high-field AMS ($\Delta K_{1/2/3}^{hf-p / hf-f}$).

Figure 5 – Synchrotron diffraction image of slate, at 0° tilt (i.e. sample slab is perpendicular to the beam). The sample's C axis (cleavage pole) is horizontal. Intensity variation along Debye rings are indicative of texture (e.g. (002) of muscovite (M002), (004) of chloritoid (Ct004) and (100) of quartz

(Q100)). This is particularly evident for the inner rings (high d-spacings).

Figure 6– Diffraction patterns along the sample's B axis (bottom) and along the sample's C axis, i.e. perpendicular to the foliation (top). Crosses indicate the measured spectrum; solid line indicates the Rietveld fit. Ticks below the spectrum show positions of diffraction peaks of contributing mineral phases (e.g. (002) and (004) of muscovite (M002; M004), (002) of chlorite (Ch002), (004) of chloritoid (Ct004) and (100) of quartz (Q100)).

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Figure 8 – Pole figures for muscovite, chloritoid and chlorite (100), (010) and (001) lattice planes (monoclinic first setting). Equal-area projection on the cleavage plane, log scale intervals in multiples of a random distribution (m.r.d.).

Figure 9 – Pole figures for quartz. Equal-area projection on the cleavage plane, linear scale intervals in multiples of a random distribution (m.r.d.).

Table headings

Table 1 – AMS parameters used in this work

Table 2 – Volume fractions and pole figure maxima and minima (in m.r.d.)

Table 3 – Magnetic tensors of the muscovite, chloritoid and chlorite single crystals, the calculated muscovite, chloritoid and chlorite phases, the measured low-field (LF) and high-field (HF) AMS and the

595 calculated 3-phase model. Abbreviations: $K_{1,2,3}$ - Eigenvalues of the magnetic tensors; K_m - bulk
596 magnetic susceptibility; T - shape parameter (Jelínek, 1978); P_J - Corrected degree of anisotropy
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Sample BR09TH131C3

Low-field AMS

	EV	Dir'n
■ K ₁	1.11	238/01
▲ K ₂	1.07	147/60
○ K ₃	0.82	328/30
P _J = 1.40 T = 0.76		

High-field para. AMS

	EV	Dir'n
◆ K ₁ ^{hf-p}	1.10	059/08
▼ K ₂ ^{hf-p}	1.07	158/50
◇ K ₃ ^{hf-p}	0.83	323/39
P _J ^{hf-p} ≥ 1.37 T ^{hf-p} ≥ 0.80		

High-field ferro. AMS

	Dir'n
✦ K ₁ ^{hf-f}	196/51
✧ K ₂ ^{hf-f}	105/01
✧ K ₃ ^{hf-f}	014/39

A, B, C. Sample axes (Cartesian coordinate system): 055/00, 145/65, 325/25

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Table 1 – AMS parameters used in this work

Property / Parameter	Equation	Reference
Bulk susceptibility (K_m)	$K_m = \frac{K_1 + K_2 + K_3}{3}$	Nagata, 1961
Corrected degree of anisotropy (P_j)	$P_j = \exp\sqrt{\{2[(\eta_1 - \eta_m)^2 + (\eta_2 - \eta_m)^2 + (\eta_3 - \eta_m)^2]\}}$ <p>with $\eta_1 = \ln K_1$; $\eta_2 = \ln K_2$; $\eta_3 = \ln K_3$; $\eta_m = \sqrt[3]{K_1 K_2 K_3}$</p>	Jelinek, 1981
Shape parameter (T)	$T = \frac{2\eta_2 - \eta_1 - \eta_3}{\eta_1 - \eta_3}$	Jelinek, 1981

Table 2 – Volume fractions and pole figure maxima and minima (in m.r.d.)

	Vol%	100		010 Qz 110		001		101		011	
Muscovite	42	4.9	0.02	5.5	0	38.6	0				
Chloritoid	13	1.8	0.5	2.6	0.3	20.9	0.5				
Chlorite	< 1	2.1	0.6	1.7	0.2	5.8	0.4				
Quartz	45			1.3	0.8	1.5	0.4	1.4	0.7	0.7	1.3

Table 3 - Magnetic tensors of the muscovite, chloritoid and chlorite single crystals, the calculated muscovite, chloritoid and chlorite phases, the measured low-field (LF) and high-field (HF) AMS and the calculated 3-phase model.

	K₁	K₂	K₃	K_m	T	P_J	k'
Chloritoid crystal	1.89E-03	1.89E-03	1.36E-03	1.73E-03	1.00	1.47	1.52E-04
Muscovite crystal	1.18E-04	1.18E-04	8.63E-05	1.08E-04	1.00	1.44	1.50E-05
Chlorite crystal	2.52E-04	2.52E-04	2.16E-04	2.40E-04	1.00	1.19	1.70E-05
Quartz	/	/	/	-1.40E-05	/	/	/
Calc. chloritoid phase	1.80E-03	1.76E-03	1.53E-03	/	0.73	1.20	6.39E-05
Calc. muscovite phase	1.16E-04	1.15E-04	9.04E-05	/	0.94	1.33	1.18E-05
Calc. chlorite phase	2.44E-04	2.41E-04	2.35E-04	/	0.39	1.04	3.85E-06
Measured AMS-LF	4.32E-04	4.17E-04	3.17E-04	3.89E-04	0.76	1.40	5.10E-05
Measured AMS-HF	4.28E-04	4.16E-04	3.22E-04	3.89E-04	0.80	1.37	4.73E-05
Calc. 3-phase tensor	2.85E-04	2.80E-04	2.39E-04	2.68E-04	0.78	1.21	2.06E-05

Abbreviations: K_{1,2,3} - Eigenvalues of the magnetic tensors; K_m - bulk magnetic susceptibility; T - shape parameter [Jelínek, 1978]; P_J - Corrected degree of anisotropy [Jelínek, 1978]; k' - magnetic anisotropy parameter for deviatoric tensors [Jelínek, 1984].