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Determination of atomic site susceptibility tensors from polarized neutron diffraction data

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Abstract

Polarized neutron diffraction provides information about the magnetization density of each individual crystallographic site. In the present paper the role of the atomic site susceptibility tensor χ_{ij} accounting for the magnetic response of individual atoms to an external magnetic field is discussed. The symmetry of this tensor is very similar to that of the tensor u_{ij} describing the thermal motion of atoms. By analogy with the atomic displacement parameters (ADPs), atomic susceptibility parameters (ASPs) can be introduced.

The six independent ASPs can be determined from polarized neutron flipping ratio measurements and visualized as *magnetic ellipsoids* which are analogous to the *thermal ellipsoids* obtained from ADPs. If the local anisotropy is small, these magnetic ellipsoids approximate to spheres with diameters proportional to the induced magnetization. In other cases, *anomalous* (elongated or flattened) ellipsoids will occur. The ASPs have been determined for the compound $\text{Nd}_{3-x}\text{S}_4$ which has the Th_3P_4 structure. They correspond to strongly oblate magnetic ellipsoids; in contrast, in the isomorphous compound Sm_3Te_4 the magnetic ellipsoids are found to be prolate.

1. Introduction

Polarized neutron diffraction is an extremely powerful tool with which to study the magnetization process in crystals. This is due to the fact that while classical magnetic measurements give information about the average magnetization, diffraction measurements provide information about the variation of the magnetization throughout the unit cell. This offers the possibility of interpreting the magnetization processes which take place in the crystal on the microscopic level. Recently a rather peculiar magnetic structure was found in the ferromagnetically ordered compounds U_3Sb_4 and U_3Bi_4 . This structure was found to contain uranium atoms of two types occupying the same crystallographic position but having different values of magnetic moment [1]. Later, a similar effect was observed in the paramagnetic compounds Sm_3Te_4 [2, 3] and $\text{Nd}_{3-x}\text{S}_4$, for which the moments induced by a magnetic field

on the rare-earth (RE) ions in equivalent sites differed considerably. This behaviour arises because, despite the overall cubic symmetry, the local symmetry of the RE ion is only tetragonal and the moments induced on each RE ion by the magnetic field depend on the orientation of the field with respect to the local tetragonal axis.

The effect can be described by attributing to each magnetic atom a site susceptibility tensor χ_{ij} which gives the magnetic response of the atom to the applied magnetic field. It is easy to show that the symmetry of the tensor χ_{ij} is the same as that of the tensor u_{ij} describing the thermal motion of atoms. The components of the tensor u_{ij} represent the mean square atomic displacement parameters (ADPs). By analogy with the ADPs, one can introduce atomic susceptibility parameters (ASPs). The response of an atom to a magnetic field can then be conveniently visualized as a *susceptibility ellipsoid* constructed from the six independent ASPs in much the same way as *thermal ellipsoids* are constructed from the ADPs.

The ASPs can be determined from polarized neutron flipping ratio measurements if the proper magnetic symmetry of the crystal is taken into account. In the absence of local anisotropy the magnetic ellipsoids reduce to spheres with their radii proportional to the induced magnetization, but in many cases *anomalous* (elongated or flattened) ellipsoids can appear. The presence of such anisotropic magnetic ellipsoids can account for the anomalous magnetic moments which have been found in two isomorphous RE compounds $\text{Nd}_{3-x}\text{S}_4$ and Sm_3Te_4 with the Th_3P_4 structure. In the body of this letter the role of the local anisotropy in determining the response of these compounds to an applied field is discussed and a formalism for calculating the magnetic scattering implicit in this model is developed. Together these allow a proper description, based on the full crystal symmetry, of the magnetization process in paramagnets containing several equivalent magnetic atoms in the unit cell.

2. Anisotropic atomic susceptibilities

The overall response of a paramagnetic material to an applied magnetic field can be described by the tensor equation

$$B_i = \sum_j m_{ij} H_j = \mu_0 \left(H_i + \sum_j \chi_{ij} H_j \right)$$

as long as the field is not so large that non-linear effects become important. With this proviso, the bulk magnetization M can be written as

$$M_i = \sum_j \chi_{ij} H_j.$$

The number of independent components of the susceptibility tensor χ_{ij} is determined by the crystal class, being one for cubic groups, two for all uniaxial groups and three, four and six for orthorhombic, monoclinic and triclinic groups respectively. In a paramagnet containing localized moments, the bulk magnetization is the vector sum of the magnetizations induced on each of the constituent magnetic atoms and these will depend on the local site symmetry, rather than the overall symmetry. A special property of magnetic diffraction is that it gives access to the wavevector-dependent magnetic response and hence to the individual site susceptibilities.

Consider a single magnetic atom (a) with an anisotropic susceptibility arising from its local environment; the magnetization induced on it by a field \mathbf{H} is given by

$$M^a = \chi^a \mathbf{H}.$$

An equivalent atom (b) related to the first by the symmetry operator $\{\tilde{\mathbf{R}} : \mathbf{t}\}$ would have magnetization

$$M^b = \chi^b \mathbf{H} = \tilde{\mathbf{R}} \chi^a \tilde{\mathbf{R}}^{-1} \mathbf{H}.$$

If the position of the first atom is \mathbf{r}_a and the distribution of moment about each atom (assumed spherical) is $\rho(\mathbf{r})$, then the magnetization distribution in the unit cell becomes

$$\mathbf{M}(\mathbf{r}) = \sum_p \tilde{\mathbf{R}}_p \chi^a \tilde{\mathbf{R}}_p^{-1} \mathbf{H} \rho(\mathbf{r} - \tilde{\mathbf{R}}_p \mathbf{r}_a - \mathbf{t}_p)$$

where the sum is over all N_g operators $\{\tilde{\mathbf{R}}_p : \mathbf{t}_p\}$ in the space group. If $f(k)$ is the magnetic form factor given by $\int_0^\infty \exp(ikr) 4\pi r^2 \rho(r) dr$, the corresponding magnetic structure factor is

$$\mathbf{M}(\mathbf{k}) = \frac{1}{N_a} f(k) \sum_p \tilde{\mathbf{R}}_p \chi^a \tilde{\mathbf{R}}_p^{-1} \mathbf{H} \exp(i\mathbf{k} \cdot (\tilde{\mathbf{R}}_p \mathbf{r}_a + \mathbf{t}_p))$$

where N_a is the number of operators q for which $\tilde{\mathbf{R}}_q \mathbf{r}_a + \mathbf{t}_q = \mathbf{r}_a$, so N_g/N_a is the multiplicity of the site a . The point group \mathcal{Q} formed by the rotational parts of the set of operators q gives the symmetry of the site a .

We recall (see for example [4]) that the scattered intensity for polarized neutrons is given by

$$I = N^2 + 2\mathbf{P}_0 \cdot (N' \mathbf{M}'_{\perp} + N'' \mathbf{M}''_{\perp}) + M_{\perp}^2,$$

where N is the nuclear structure factor with real and imaginary parts N' and N'' , \mathbf{M}'_{\perp} and \mathbf{M}''_{\perp} are the real and imaginary parts of the magnetic interaction vector \mathbf{M}_{\perp} defined by $\mathbf{M}_{\perp}(\mathbf{k}) = \mathbf{k} \times \mathbf{M}(\mathbf{k}) \times \mathbf{k}$ and \mathbf{P}_0 is the neutron polarization vector. The quantity measured experimentally is the *flipping* ratio, $R = I^+/I^-$, between the intensity of neutrons scattered when polarized parallel (I^+) and anti-parallel (I^-) to the external magnetic field. R can easily be expressed in terms of the susceptibility parameters using the equations given above and hence the flipping ratios can be used to determine these parameters using least-squares refinement techniques.

3. Symmetry constraints on the site susceptibility

Using the property of symmetry rotations that $R_{ij} = R_{ji}^{-1}$, the symmetry constraints on the components of the site susceptibility tensor are given by the set of equations

$$\chi_{ij} = \sum_{k=1}^{k=3} \sum_{l=1}^{l=3} R_{ik} R_{jl} \chi_{kl} \quad \text{for } 1 < i \leq 3; 1 < j \leq 3$$

for all operators $\tilde{\mathbf{R}}$ in the group \mathcal{Q} . These are the same symmetry constraints as apply to other quantities which relate two vectors, such as the anisotropic thermal vibration parameters. There are at most six independent components of the symmetric tensor χ_{ij} ; they can be labelled with the single index m with $m = 1; 2; 3$ for $ij = 11; 22; 33$ and $m = 4; 5; 6$ for $ij = 23, 32; 31, 13; 12, 21$. To include determination of these components in a least-squares refinement procedure it is convenient to pre-calculate the set of 6×3 matrices $\tilde{\mathbf{P}}$ where

$$P_{im} = \sum_{j=1}^{j=3} R_{ik} R_{jk} h_j \quad \text{for } m = 1, 2, 3 \quad (k = 1, 2, 3)$$

and

$$P_{im} = \sum_{j=1}^{j=3} (R_{ik} R_{jl} + R_{il} R_{jk}) h_j \quad \text{for } m = 4, k = 2, l = 3$$

$$m = 5, k = 3, l = 1$$

$$m = 6, k = 1, l = 2$$

where the h_j are the direction cosines of the magnetizing field. The moment induced by a field of strength H on any atom related to the representative atom by the operator p can then be written as

$$M_i = H \sum_{m=1}^{m=6} P_{im} \chi_m$$

and the derivatives of the magnetic structure factors with respect to the components of the susceptibility tensor are given by

$$\frac{\partial M_i(k)}{\partial \chi_m} = \frac{H}{N_a} f(k) \sum_p P_{im} \exp(i\mathbf{k} \cdot (\tilde{\mathbf{R}}_p \mathbf{r}_a + \mathbf{t}_p))$$

a form which can easily be introduced into a least-squares program.

4. Experiment

Neutron diffraction measurements were carried out at the reactor ORPHÉE, LLB CEA, Saclay. Prior to the polarized neutron study the crystal of $\text{Nd}_{3-x}\text{S}_4$ of dimensions $1.5 \times 1.5 \times 1.5 \text{ mm}^3$ was characterized using the four-circle neutron diffractometer 6T2 with neutrons of wavelength $\lambda = 0.9 \text{ \AA}$. Integrated intensities of 150 reflections were measured at room temperature and were used to refine the single adjustable positional parameter, X , of sulphur, the site occupation factor of Nd, the isotropic temperature factors and the extinction parameters. The latter were found to be insignificant. The refinement gave $X = 0.0735$ for the sulphur position and 0.88 for the Nd site occupancy.

Polarized neutron measurements were performed on the two-axis lifting-counter diffractometer 5C1 using a wavelength $\lambda = 0.845 \text{ \AA}$ (Heusler alloy monochromator). The polarization of the incident neutron beam was $P_0 = 0.91$. Higher-order contamination was suppressed to less than 0.01% by means of erbium filters. The temperature dependence of the magnetic susceptibility was measured directly by means of polarized neutron scattering. For this purpose the spin-up (I^+) and spin-down (I^-) intensities of the 211 reflection were measured as a function of temperature and the difference $I^+ - I^-$ plotted versus temperature (see figure 1). This difference,

$$I^+ - I^- = 2\chi H F_N (P_0^+ - P_0^-),$$

is proportional to the Fourier component (211) of the wavevector-dependent susceptibility of the crystal. As seen from the figure, $\text{Nd}_{3-x}\text{S}_4$ exhibits Curie–Weiss behaviour with $\theta_p \approx -15 \text{ K}$ but remains paramagnetic down to 1.5 K. Two sets of polarized neutron flipping ratios were measured at 2 K. In the first a vertical magnetic field of 7 T was applied nearly parallel to the cubic [001] axis of the crystal. In fact there was a misalignment of about 10° between the c -axis and the magnetic field direction. (The exact orientation of the magnetic field direction with respect to the cubic crystal axes was taken properly into account in the final data analysis.) A total of 116 flipping ratios with $\sin \theta / \lambda < 0.4 \text{ \AA}^{-1}$ were measured. Because of the misalignment mentioned above, the flipping ratios of symmetry-equivalent reflections were not averaged together.

The second set consisted of 126 flipping ratios also measured at 2 K but with the magnetic field not along any of the symmetry directions of the cubic structure. The orientation of the field in this case was close to the [421] direction.

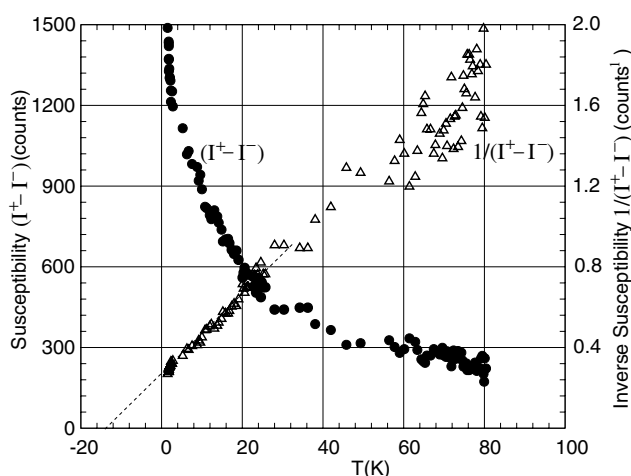


Figure 1. The magnetic susceptibility (full circles) and inverse susceptibility (open triangles) of $\text{Nd}_{3-x}\text{S}_4$ deduced from the difference in intensity of neutrons scattered by the 211 reflection with polarization parallel (I^+) and anti-parallel (I^-) to the applied field.

5. Results and discussion

In $\text{Nd}_{3-x}\text{S}_4$ (cubic space group $I\bar{4}3d$) twelve Nd atoms occupy a single site (12a) with local symmetry $\bar{4}$. These twelve magnetic atoms can be subdivided into three different groups which have their local tetragonal axes along x , y and z respectively. As has been shown in [3], when a magnetic field is applied parallel to one of the cubic fourfold axes the low-symmetry (tetragonal) $I\bar{4}2d$ group has to be used in the magnetic moment refinement. This is the highest-symmetry subgroup of the paramagnetic group $I\bar{4}3d$ under which the homogeneous magnetization component M_Z , induced parallel to the fourfold axis by the magnetic field, is invariant. In this subgroup the 12a sites of the cubic group split into two different subsets; one fourfold one (Nd1) in which the field axis is parallel to the local tetrad and an eightfold one (Nd2) in which it is perpendicular. The refinement of the first data set using the tetragonal group $I\bar{4}2d$ demonstrated clearly that there is a very large difference between the magnetic moments induced parallel and perpendicular to the local tetragonal axes. The moments induced on those Nd atoms whose tetrad axes were parallel to the field were $\mu_{\text{Nd1}} = 0.62(5) \mu_B$ compared to $\mu_{\text{Nd2}} = 1.44(5) \mu_B$ for those with the perpendicular orientation as illustrated in figure 2. The goodness of fit $\chi^2 = 5.14$ for 116 reflections. For comparison, a model in which the local anisotropy was neglected and all Nd atoms have the same moment gives $\mu_{\text{Nd}} = 1.20(5) \mu_B$ and $\chi^2 = 22.5$.

There are severe difficulties in generalizing this method of analysing the data since, for different field orientations, different subgroups of the original paramagnetic group ($I\bar{4}3d$) must be used. The set of magnetic atoms which are equivalent in the absence of magnetic field has to be split into several new sets which, because of their anisotropy, could have different induced magnetic moments. Thus the choice of the subgroup compatible with the homogeneous magnetization component induced by the field, the corresponding sets of non-equivalent atoms and consequently the number of independent variables depend on the orientation of the applied magnetic field. This becomes clear in considering how to treat the set of flipping ratios measured with the field applied along the [421] direction. In this case only a triclinic space group is compatible with the induced homogeneous magnetization, which

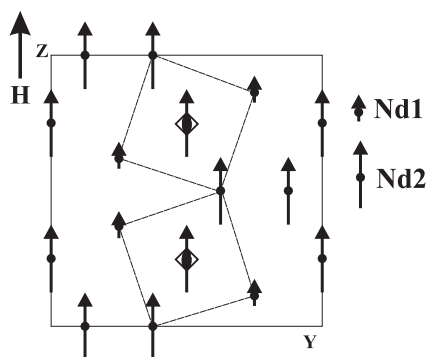


Figure 2. Magnetic moments induced in $\text{Nd}_{3-x}\text{S}_4$ at 2 K, by a field $H = 7$ T parallel to [001].

implies that all six moments in the primitive unit cell can have different moments and should be varied independently.

These difficulties can be overcome using the concept of site susceptibility. In this approach the point group symmetries of the atomic sites of the paramagnetic group $I\bar{4}3d$ are used to constrain the site susceptibility tensors. This allows the magnetic moments induced on different atoms by a magnetic field of arbitrary orientation to be described using the full symmetry of the paramagnetic group. The symmetry constraints on the susceptibility tensor for a magnetic atom occupying the 12a site in the group $I\bar{4}3d$ imply $\chi_{11} = \chi_{22} \neq \chi_{33}$ and $\chi_{12} = \chi_{13} = \chi_{23} = 0$. Thus only two independent susceptibility parameters need to be determined regardless of the field direction.

The two independent components of susceptibility were determined from a least-squares fit of the site susceptibility model to the flipping ratios using a least-squares refinement procedure incorporating the results given in section 2 and built from routines in the CCSL [5, 6]. It may be noted that the magnetic structure induced by the magnetic field from the site susceptibilities is not in general collinear. The non-collinearity is taken into account in the magnetic structure factor calculation on which the flipping ratio refinement is based. The refinement carried out with the 116 flipping ratios measured with the magnetic field $H = 7$ T applied parallel to [001] gave $\chi_{11}H = \chi_{22}H = 1.45(5) \mu_B$ and $\chi_{33}H = 0.55(5) \mu_B$ with the goodness of fit $\chi^2 = 4.4$. For convenience, the results are given as induced magnetic moments (the susceptibility components multiplied by the magnetic field), since this allows easy comparison with the results of a conventional least-squares refinement based on the localized magnetic moment model. It is clear, however, that the χ_{ij} can easily be transformed to more conventional units.

This analysis demonstrates that the susceptibility (and consequently the magnetization) along the local tetragonal axis is nearly three times less than that perpendicular to the axis. The same refinement procedure applied to the second data set of 122 flipping ratios measured with the field parallel to the [421] direction gave $\chi_{11}H = \chi_{22}H = 1.43(8) \mu_B$ and $\chi_{33}H = 0.76(16) \mu_B$ with $\chi^2 = 3.9$. These parameters are the same, within experimental error, as those obtained with the field along [001]. It should be pointed out that the relatively poor precision in the determination of χ_{33} from the second set of data is due to rather strong correlation (81%) between the parameters χ_{11} and χ_{33} . These results confirm that the polarized neutron data collected with different field orientations can be analysed within the paramagnetic group $I\bar{4}3d$ using just the two susceptibility parameters allowed by symmetry.

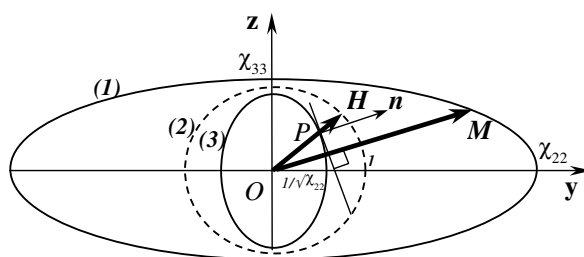


Figure 3. A principal section through (1) the magnetic ellipsoid, (2) a unit sphere and (3) the representation quadric for susceptibility.

The relationship between the site and bulk susceptibility tensors is quite straightforward:

$$\chi^b = \sum_m \sum_q \mathbf{R}_q \chi^{mq}$$

where χ^{mq} is the susceptibility tensor of the q th equivalent position (that generated by the symmetry operator whose rotational part is \mathbf{R}_q) for the m th independent magnetic atom. The symmetry properties of the tensors ensure that the bulk susceptibility tensor transforms with the full symmetry of the crystallographic class. For cubic $\text{Nd}_{3-x}\text{S}_4$ the bulk susceptibility is isotropic and the bulk susceptibility tensor contains the single independent element $\chi_{11}^b = \chi_{22}^b = \chi_{33}^b$. Using the symmetry properties of the site susceptibility tensors of the twelve Nd atoms in the unit cell, the above equation reduces to

$$\chi_{11}^b = 4(2\chi_{11} + \chi_{33}).$$

This expression gives the full bulk susceptibility as long as the contribution from sulphur atoms and the conduction electrons can be neglected. Since the diffraction data fitted well with the model based on atomic U moments, the sulphur contribution can probably be neglected. On the other hand the conduction electron contribution cannot be determined from the neutron experiment.

Once the site susceptibility parameters are known, one can easily calculate the magnitude and the direction of the moment induced on each Nd atom within the unit cell by a magnetic field applied in an arbitrary direction, since the site susceptibility tensor relates the vectors \mathbf{M}^a and \mathbf{H} .

It is often useful to represent the properties of a second-rank tensor by a quadratic surface constructed from its independent components [7]. This surface gives a complete representation of the tensor and can be used to derive any crystal property to which it relates. Such a surface can be constructed from the site susceptibility tensor in two different ways. One representation is given by the equation

$$\frac{x^2}{\chi_{11}^2} + \frac{y^2}{\chi_{22}^2} + \frac{z^2}{\chi_{33}^2} = 1$$

in which the axes are the principal axes of χ_{ij} (note that in the present case the principal axes of the tensor coincide with the main crystal axes). It is an ellipsoid, whose semi-axes are χ_{11} , χ_{22} and χ_{33} (figure 3). This surface has the property that its radius vectors \mathbf{M} give the magnitude and direction of the magnetization which can be induced by a field \mathbf{H} of fixed (unit) magnitude and arbitrary direction. This surface will be called the magnetic ellipsoid. Unfortunately it does not give the relationship between the directions of \mathbf{M} and \mathbf{H} in a simple

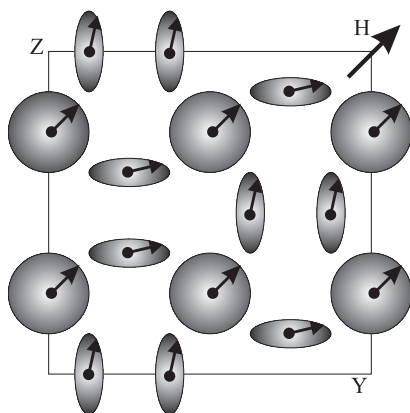


Figure 4. The [100] projection of the unit cell of $\text{Nd}_{3-x}\text{S}_4$ showing the magnetic ellipsoids at 2 K for $\mathbf{H} \parallel [011]$.

way. This relationship is more easily derived from the alternative representation surface (the representation quadric for susceptibility):

$$\chi_{11}x^2 + \chi_{22}y^2 + \chi_{33}z^2 = 1$$

which is an ellipsoid, whose semi-axes are $1/\sqrt{\chi_{11}}$, $1/\sqrt{\chi_{22}}$ and $1/\sqrt{\chi_{33}}$. The direction of M^a is the normal to the surface of the representation quadric at the point 'P' (figure 3) at which the radius vector 'OP' parallel to \mathbf{H} intersects the surface [7].

The geometrical interpretation of the magnetic ellipsoid can be summarized as follows. When \mathbf{H} is directed along any of the three principal axes of the ellipsoid, M^a is parallel to \mathbf{H} , but the induced moment is different for different axes. When \mathbf{H} is not parallel to a principal axis, M^a is no longer parallel to \mathbf{H} , but has components both parallel and transverse to it. For the case of Nd_3S_4 the local tetrad axis may be thought of as a *direction of hard magnetization* and M^a has a tendency to turn away from it. Note that polarized neutrons give access to only the parallel component of the induced moment, but the transverse (non-collinear) parts of the moments could also be estimated from neutron diffraction data by measuring the total integrated intensity of the magnetic scattering, rather than just the polarization-dependent part.

Magnetic ellipsoids representing the site susceptibilities of all the Nd atoms in Nd_3S_4 are shown in figure 4. As seen from the figure, the strongly flattened ellipsoids (oblate susceptibility tensor) occur in three different orientations, each having its short axis parallel to the tetrad axis on which it lies. The arrows on the figure represent the magnitudes of the Nd moments which are induced by a field applied parallel to the [011] direction, calculated using the refined ASPs. It can be seen that only one third of the Nd moments lie parallel to the field, while the rest have quite different directions. This rather complex non-collinear distribution of magnetic moments is not however incompatible with the overall cubic symmetry of the crystal, since the bulk magnetization, given by the vector sum of all magnetic moments, remains parallel to the field and invariant with respect to the field orientation.

It is interesting to compare this result with the result reported earlier for the isomorphous compound Sm_3Te_4 [3] in which Sm ions occupy the same crystallographic positions as Nd in $\text{Nd}_{3-x}\text{S}_4$. According to [3], the moment induced along the local tetragonal axis of the Sm ions was found to be about five times higher than that induced perpendicular to it, in sharp contrast to the case for the Nd compound. Thus Sm_3Te_4 represents a case of strongly elongated (*prolate*) local susceptibility.

As has been mentioned above, the symmetry properties of tensors χ_{ij} and u_{ij} are the same. It is interesting to note several similarities between ADPs and atomic susceptibility parameters (ASPs):

- (i) ADPs are usually considered as a probe of the shape of the atomic potential well. The ASPs can be considered as a probe of the magnetic Hamiltonian.
- (ii) ADPs give information about atomic vibration, dynamics, disorder. ASPs give information about magnetic correlation and spin dynamics.
- (iii) Symmetry constraints for ASPs are the same as those for ADPs.
- (iv) Anomalous ADPs can indicate the channel for structural phase transitions; similarly the ASPs can suggest the way in which the crystal may order magnetically. This latter point is well illustrated by the magnetic structure of $U_3Si_2Al_3$ which is discussed in detail in the following paper [8].

6. Conclusions

It has been shown that anisotropy due to the local environment of magnetic atoms can give rise to the appearance of different magnetic moments on equivalent crystallographic positions when magnetization is induced in the paramagnetic region by an applied magnetic field. Such anisotropy can be described by site susceptibility tensors, the number of independent components of which can vary from 2 for uniaxial site symmetries to 6 for triclinic sites. The magnitude and direction of the moment induced on each particular atom in the unit cell depends on the orientation of the principal axes of the susceptibility tensor with respect to the field. Polarized neutron scattering can be used as a microscopic probe of such local anisotropy. Nd_3S_4 and Sm_3Te_4 represent two extreme types of local anisotropy existing for the crystallographic sites with uniaxial (tetragonal) symmetry, which can be called the *prolate* and the *oblate* susceptibilities.

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