Extensive studies on the low-temperature properties of $TbPO_4$. II. Measurements of the Faraday rotation

Extensive studies on the low-temperature properties of TbPO$_4$: II. Measurements of the Faraday rotation

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Abstract. Studies of the Faraday rotation in TbPO$_4$ confirmed the known properties of the different low-temperature phases. Unexpected oscillations, however, were observed in the temperature dependence of the measured rotation in one of the low-symmetry phases. Model calculations of the rotation for a material possessing both Faraday rotation and birefringence did allow an interpretation of the findings, but rather unusual properties of the substance have to be assumed. These calculations additionally showed that the method of measurement influences the results in a typical way so that the optical properties of the substance cannot always be derived directly from the experimental curves.

1. Introduction

In the series of rare-earth compounds with zircon structure (space group $I4_1/amd$, site symmetry $\overline{4}m2$) there are only a few that undergo a structural as well as a magnetic phase transition at low temperatures. In the vanadates and arsenates of dysprosium and terbium, the cooperative Jahn–Teller interaction is stronger or even much stronger than the magnetic interactions, thus the structural transition occurs first (Gehring and Gehring 1975). In TbPO$_4$, however, these two interactions are of almost comparable size with the consequence that first of all a simple antiferromagnet (AF) with moments along the crystal c axis and tetragonal symmetry is formed below $T_{N1} = 2.28 \pm 0.02$ K. Shortly afterwards, a canted antiferromagnet (AF′) with the moments tilted off the c axis in a {110} plane (still collinear) and monoclinic symmetry is generated below $T_{N2} = 2.13 \pm 0.02$ K. An external magnetic field along c forces the crystal into a spin-flop-like phase SF before it enters the saturated paramagnetic phase PM.

In the preceding paper (Mensinger *et al* 1992), henceforth referred to as I, magnetic measurements on TbPO$_4$ were described. These were directed, on the one hand, towards the determination of the phase boundaries and the magnetic properties of the different phases and, on the other hand, towards the search for hints of additional phase transitions found in the birefringence measurements of the following paper (Müller *et al* 1992), to be referred to as III.

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In the present paper we report on measurements of the Faraday rotation that we have undertaken. Our expectation was not only to confirm the results of our magnetic measurements but also to get further information in particular on the low-symmetry phases since for these a contribution of the small but not insignificant birefringence will be superimposed on the Faraday rotation. This latter expectation, however, could not be performed. On the other hand, we have observed an interesting oscillation of the measured rotation that we would like to make known although our attempts to explain it are not completely satisfactory.

2. Experimental details

The flux-grown tetragonal crystals of TbPO$_4$ (Hintzmann and Müller-Vogt 1969) were of good optical quality. Samples from different growth batches showed the same results within the accuracy of measurement. This paper is concentrated on the findings on one crystal only. Its dimensions are $a \times b \times c = 1.03 \times 2.35 \times 2.45$ mm^3, its approximate demagnetizing factor in the c direction is $N_c \approx 0.20$ (SI units). Since below T_{N2} the substance is distorted in a $\{110\}$ plane its properties are described either in the crystallographic a, b, c system or in an x, y, z system which is rotated by 45° around the $c \equiv z$ axis.

To measure the Faraday rotation, the crystal and a piezoelectric modulator (50 kHz) with modulation in the x direction were placed between two linear polarizers. The transmission direction of the first polarizer was set at 45° with respect to the x direction. The sample was irradiated with monochromatic light of a He–Ne laser ($\lambda = 632.8$ nm) parallel to the c direction. To reduce its intensity a grey filter and a pinhole in front of the crystal were used. The rotation angle α_0 was measured by computer-controlled turning of the analyser using the lock-in technique. A resolution in α_0 of 3×10^{-3} rad was obtained.

An exchange-gas cryostat with optical windows for a horizontal light path and a superconducting split-coil magnet were used. The phase difference produced by the birefringence of the windows was measured to be smaller than 2×10^{-2} rad. Hence it could be neglected against the birefringence (about 0.6 rad) and the rotation (about 9 rad at an internal field of 0.5 T) of the sample. Temperatures down to 1.4 K (with absolute accuracy ±20 mK, short-time stability < 1 mK) and magnetic fields up to 4.1 T (with absolute accuracy ±1.5%, short-time stability < 0.1 mT) were available.

3. Experimental results

Faraday rotation means the rotation of the polarization direction of linearly polarized light passing through the magnetized sample parallel to the magnetization direction. Hence, the rotation was measured with the external field along c either as a function of temperature at constant field (temperature sweeps) or as a function of field at constant temperature (field sweeps). In this orientation, all domains are magnetically equivalent, thus no change of the domain structure caused by the field is to be expected.

Both the temperature and field sweeps show a great similarity with the corresponding curves measured for the magnetization (see paper I). Hence, they will not be reproduced here. Within the accuracy of the measurement, the curves

are proportional to each other for temperatures above T_{N2} and for fields above 1.1 T, i.e. in the range of the AF and PM phases. Larger differences are clearly discernible in the AF′ phase near to the AF′–AF phase boundary. At the beginning of the jump there is no steady turn in the field sweeps but a sharp bend and the jump itself is steeper in the rotation than in the magnetization. Demagnetization does not transform the jump into a vertical step but into an increase with overhanging to the left. This may be caused by the inhomogeneity of the demagnetizing field in a crystal of non-ellipsoidal shape, especially in its central cylinder of small diameter that has been used for the measurements, see Brug and Wolf (1985). Slight differences are detectable at low fields in the AF′ phase. However, contrary to our expectation, no significant differences could be discerned in the field sweeps within the SF phase. The distinguishable differences are so small that an analysis seems senseless considering the accuracy of measurement of 5% for the entire quantity.

The temperature and field values of the phase transitions were determined in the same manner as in paper I. They are included in the phase diagram of figure 5 of paper III and are in very good agreement with our former measurements, despite lower accuracy due to difficulties in the demagnetization process.

An unexpected behaviour was observed, however, in the temperature dependence of the Faraday rotation. For external fields in the range from 0.6 to 0.9 T, the measured rotation oscillates with small amplitude (largest amplitude of about 0.2% at $B_{ext} = 0.7$ T). The period in temperature increases with field strength in such a way that backward extrapolation would give zero period for zero field. A typical example is shown in figure 1(*a*). The oscillations are reproducible and are not influenced by the time constant of the equipment. They are independent of the particular part of the crystal (centre or periphery) that is used for the experiment. And they do not change when the crystal is rotated around the *c* axis or when polarizer, modulator and analyser are rotated simultaneously. Some results obtained under these different experimental conditions are shown in the curves of figure 1(*b*). Corresponding oscillations were found neither in our magnetization nor in our magnetic susceptibility measurements.

These oscillations appear in the range of the spin-flop-like SF phase for which the magnetic structure and crystal symmetry are not yet known. Following Kasten and Üffinger (1985), for this phase one can presume that the moments of both of the two sublattices are symmetrically tilted off the *c* axis with equal components in the field direction and that the tilt angle will decrease with increasing field, going to zero at the SF–PM phase boundary. Thus, including the magnetic symmetry, the crystal will have monoclinic symmetry with a distortion expected to be in the same direction as found for the AF′ phase, i.e. in the *x* direction for the one type and in the *y* direction for the other type of domain. The distortion of the crystal causes a birefringence. The tilt angle of the moments will change not only with field but also with temperature. And a change of the tilt angle will result in a change of the *z* component of the magnetization (and hence the Faraday rotation) as well as in a change of the transverse component (and hence the distortion and the birefringence). These considerations could lead to the assumption that the observed oscillations are effected either by a monotonic change of the Faraday rotation at essentially constant birefringence or a monotonic change of the birefringence at essentially constant Faraday rotation or a monotonic change of both quantities. This will be examined in section 4. Since the deviation of the monoclinic crystal symmetry from an orthorhombic one will be small and since the calculations of paper III for this phase yield simply an orthorhombic distortion,
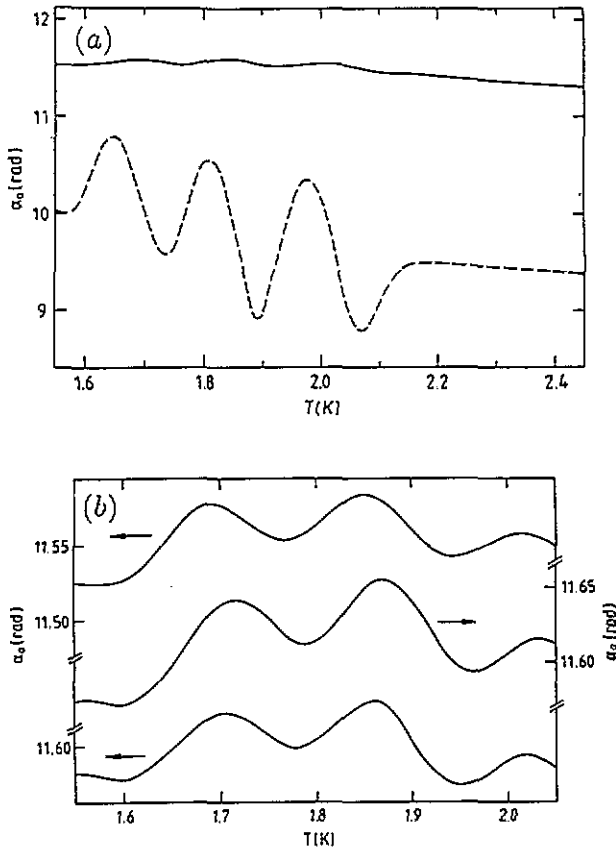
Figure 1. Temperature dependence of the rotation angle α_0 measured at an external field of 0.7 T. Averaged curves are shown. The scattering of the measuring points lies within the width of the full line in (*a*), it is twice the width of the full lines in (*b*). (*a*) Experimental curve (full line) and its derivative $d\alpha_0/dT$ (broken line, not to scale). (*b*) Curves for different experimental conditions in an enlarged scale. The upper curve is measured at the centre of the crystal; the middle curve is measured near the outer surface of the crystal and the lower curve is measured at the centre of the crystal after turning the polarizer, modulator and analyser by 45°. The small differences in the absolute values of the curves will most probably be caused by slightly differing field strengths in the different sweeps.

the latter symmetry will be assumed.

We shall see that the assumption of a monotonic change in one or two of the quantities is too simple and thus not capable of explaining the observed oscillations (see section 4.2). The following calculations will, however, indicate which properties ought to be assigned to TbPO$_4$ to bring the result of the calculation into agreement with experiment. Additionally, we would like to demonstrate, in general, that uncommon effects may be observed in rotation measurements on a substance having both Faraday rotation and birefringence, for example, that oscillations or more or less pronounced steps may appear even if Faraday rotation or birefringence or both of them are varying regularly (see section 4.3).

4. Theoretical interpretation of measurements on a material possessing both Faraday rotation and birefringence

4.1. Derivation in general

If light is propagating along the z axis of an orthorhombic crystal having both Faraday rotation and birefringence, it is split into two orthogonal elliptically polarized normal modes. Their phase difference φ after passing through the sample of thickness d is given by

$$\varphi = d\sqrt{\rho^2 + (2\vartheta)^2} \tag{1}$$

where ρ (intrinsic birefringence) or 2ϑ (intrinsic Faraday rotation) are the phase differences per unit length of the material if it exhibited no Faraday rotation or no birefringence, respectively. It is convenient to use the system of the x, y crystal axes. Then the electric vectors of the light at the front ($z = 0$) and the end ($z = d$) surfaces of the sample are related in the following way (see, for example, Hecht 1987)

$$\begin{pmatrix} E_x \\ E_y \end{pmatrix}_{z=d} = \mathcal{J}_{\text{cryst}} \begin{pmatrix} E_x \\ E_y \end{pmatrix}_{z=0}. \tag{2}$$

Here

$$\mathcal{J}_{\text{cryst}} = \begin{pmatrix} \cos(\varphi/2) - \mathrm{i}\cos\chi\sin(\varphi/2) & -\sin\chi\sin(\varphi/2) \\ \sin\chi\sin(\varphi/2) & \cos(\varphi/2) + \mathrm{i}\cos\chi\sin(\varphi/2) \end{pmatrix} \tag{3}$$

is the Jones matrix of the crystal (Tabor *et al* 1969 and 1970) and the angle χ (strictly speaking, $\tan\chi$) relates the relative strength of the Faraday rotation to that of the birefringence with

$$\sin\chi = 2\vartheta/\sqrt{\rho^2 + (2\vartheta)^2} \qquad \cos\chi = \rho/\sqrt{\rho^2 + (2\vartheta)^2}. \tag{4}$$

We will first discuss the result for the normal mounting of the optical elements as described in section 2. The polarizer set at 45° off the x direction leads to equal amplitude $(E_x)_0 = (E_y)_0$ along x and y. The Jones matrix of the modulator with modulation in the x direction and that of the analyser are

$$\mathcal{J}_{\text{mod}} = \begin{pmatrix} e^{\mathrm{i}\Gamma} & 0 \\ 0 & 1 \end{pmatrix} \qquad \text{and} \qquad \mathcal{J}_{\text{an}} = \begin{pmatrix} \cos^2\alpha & \sin\alpha\cos\alpha \\ \sin\alpha\cos\alpha & \sin^2\alpha \end{pmatrix} \tag{5}$$

where $\Gamma = \Gamma_0\cos\omega t$, $\omega/2\pi = 50$ kHz and α = angular position of the analyser. Then the amplitudes at the photoelectric cell are given by

$$\begin{pmatrix} E_x \\ E_y \end{pmatrix}_{\text{phot}} = \mathcal{J}_{\text{anal}}\mathcal{J}_{\text{cryst}}\mathcal{J}_{\text{mod}} \begin{pmatrix} E_x \\ E_y \end{pmatrix}_{z=0} \tag{6}$$

and the intensity

$$I \sim (|E_x|^2_{\text{phot}} + |E_y|^2_{\text{phot}}). \tag{7}$$

Of the series expansion of the intensity in multiples of ω, $I = I_0 + I_\omega + I_{2\omega} + \ldots$, only the second harmonic

$$
\begin{aligned}
I_{2\omega} &\sim -\sin\chi\sin\varphi\cos 2\alpha + \cos\varphi\sin 2\alpha \\
&= A\sin(2\alpha - 2\alpha_0)
\end{aligned}
\tag{8}
$$

is amplified by the lock-in technique. Here α_0 is the angular position of the analyser for zero intensity $I_{2\omega}$, i.e. the measured rotation angle for the crystal of thickness d, with

$$
\tan 2\alpha_0 = \sin\chi\tan\varphi.
\tag{9}
$$

The amplitude, $A = \sqrt{\sin^2\chi\sin^2\varphi + \cos^2\varphi}$, is a measure of the change in intensity $I_{2\omega}$ with increasing distance $(\alpha - \alpha_0)$ from the zero position. Introducing (1) and (4) into equation (9), one gets

$$
\alpha_0 = \tfrac{1}{2}\arctan\left\{\left(2\vartheta/\sqrt{\rho^2 + (2\vartheta)^2}\right)\tan\left(d\sqrt{\rho^2 + (2\vartheta)^2}\right)\right\}.
\tag{10}
$$

Obviously, there is no simple relation between the measured rotation angle α_0 and the phase difference φ, or the birefringence ρ, the Faraday rotation 2ϑ and the thickness d, respectively. Inspection of this equation leads to the following conclusions.

(i) For constant birefringence (fixed ρ), the rotation angle α_0 is increasing with growing Faraday rotation in a monotonic way without any steps, the difference between $2\alpha_0$ and $2\vartheta d$ becoming smaller as 2ϑ increases.

(ii) For constant Faraday rotation (fixed 2ϑ), on the other hand, the rotation angle α_0 is a step function for growing birefringence with steps at $\varphi = d\sqrt{\rho^2 + (2\vartheta)^2} = (2N + 1)\pi/2$ and with plateaux at $\alpha_0 = N'\pi/2$ for integer N and N'. Because of the decreasing factor in front of the tangent on the right-hand side of equation (10), the steps are getting steeper and the plateaux more pronounced for increasing ρ. The calculated curve of the rotation angle α_0 for fixed $2\vartheta d = 8.55$ rad is shown in figure 2.

(iii) For simultaneous variation of 2ϑ and ρ, including increasing 2ϑ and decreasing ρ or vice versa, the steps in α_0 are less distinct.

Next we will discuss the result for the case when the crystal, i.e. the x, y axes are rotated by 45° in comparison with those of the normal mounting as handled before. The transmission direction of the polarizer is along x, the modulation direction of the modulator is 45° to x. Using the same procedure as in the former case, one obtains, after some lengthy calculation, the rotation angle

$$
\alpha_0' = \tfrac{1}{2}\arctan C
\tag{11}
$$

with (φ as defined in equation (1))

$$
C = 4\vartheta d(\sin\varphi/\varphi)/[(1 + \cos\varphi) + (1 - \cos\varphi)(\rho^2 - (2\vartheta)^2)/(\rho^2 + (2\vartheta)^2)].
\tag{12}
$$

Now the following conclusions can be drawn.

(i) For constant birefringence ρ, the rotation angle α_0' is monotonically increasing with growing Faraday rotation 2ϑ, as soon as $2\vartheta \approx \rho$.

(ii) For constant Faraday rotation 2ϑ, on the contrary, the rotation angle α_0', mostly after a starting increase, shows a periodic oscillation with decreasing amplitude as a function of birefringence ρ, as soon as $\rho > 2\vartheta$. The oscillations are mainly given by the sine term in the numerator of (12) (which determines the period of the oscillations) whilst the denominator goes essentially to unity for large ρ. However, it is very important that the oscillations occur around special angles which are $\alpha_0' = N'\pi$ and that these values of α_0' are valid for $\varphi = N\pi$ with integer N' and N. The curve of the rotation angle α_0' calculated for fixed $2\vartheta d = 8.55$ rad is reproduced in figure 3.

(iii) Simultaneous variation of 2ϑ and ρ shows either a monotonically increasing or oscillating rotation angle α_0', essentially depending on whether $2\vartheta > \rho$ or $2\vartheta < \rho$. The values of the special angles are the same as in (ii).
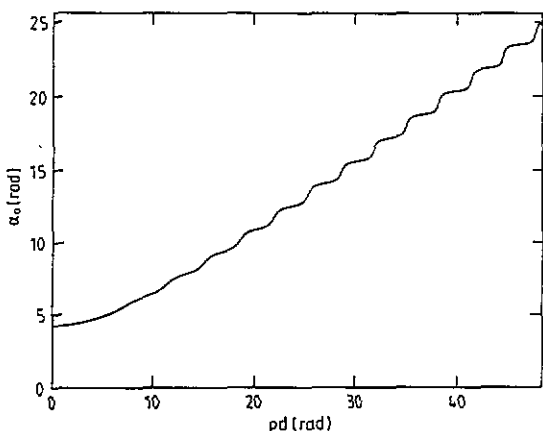

Figure 2. Rotation angle α_0 calculated according to equation (10) for increasing birefringence ρd and constant Faraday rotation $2\vartheta d = 8.55$ rad.

4.2. Application to TbPO$_4$

After these general considerations, the strength of the Faraday rotation and the birefringence that might be on hand in TbPO$_4$ shall now be estimated. The value of α_0 measured at 1.75 K for an internal field $B_{int} = 0.47$ T in the low-field region of the SF phase is 8.58 rad. In paper III the birefringence of a single-domain crystal at zero field, i.e. in the AF$'$ phase, is measured as $\delta n_{yx} = \lambda\rho/2\pi = 25 \times 10^{-6}$ for $\lambda = 632.8$ nm, thus $\rho d = 0.61$ rad ($d = 2.45$ mm). Using this as a rough value of the upper limit for the SF phase, equation (10) leads to $2\vartheta d = 8.55$ rad. Hence, in the SF phase the Faraday rotation is about 15 times larger than the birefringence. In the curves of figures 2 and 3, step-wise increase or oscillations are pronounced only for $\rho > 2\vartheta$. This relation is obviously not fulfilled in TbPO$_4$. In the range where ρ is very small against 2ϑ the curves of figures 2 and 3 are steady.

The reader may doubt that it is justified to set the value of the birefringence measured at zero field in the AF$'$ phase as an approximate upper limit for the SF phase. A direct determination from the measured rotation in the SF phase requires,
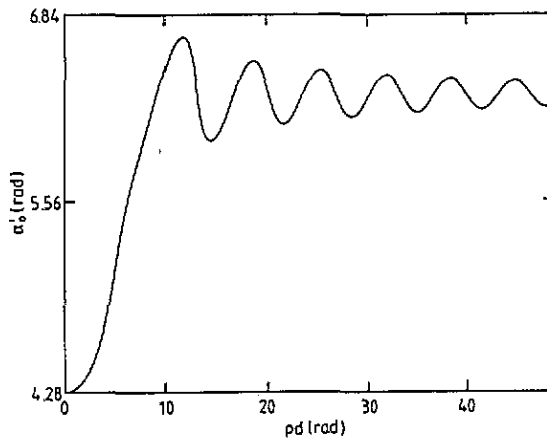
Figure 3. Rotation angle α_0' calculated according to equations (11) and (12) for increasing birefringence ρd and constant Faraday rotation $2\vartheta d = 8.55$ rad.

however, the knowledge of the (unknown) Faraday rotation. But the calculations performed in paper III lead to two important results regarding this matter:

(i) There is obviously a proportionality between the calculated orthorhombic distortion of the crystal and the measured birefringence.

(ii) The orthorhombic distortion increases in the AF′ phase for increasing field along c, but only by a factor of about two from zero field to the AF′–SF boundary (this being the highest value at a fixed temperature in the entire phase diagram for this field direction). After that the distortion quickly decreases to very small values at the SF–PM boundary. Doubling the upper limit of the birefringence for the SF phase, leads to a value of $2\vartheta d = 8.49$ instead of 8.55 for the Faraday rotation, but the gross relation $\rho < 2\vartheta$ is maintained.

Another objection may be related to the point whether the crystal, used to measure the birefringence at zero field (see paper III), was really single-domain. There are many reasons to believe that this is so. Nevertheless, it is not absolutely out of the question that the crystal was multi-domain, but with a very special domain structure that (for example, owing to defects in the crystal) was reproduced even after diverse cycles of increasing and decreasing field strengths. Since the birefringence of a single domain with distortion in x and of one with distortion in y direction are equal, but of different signs, the measured birefringence is given by the birefringence of an actual single-domain crystal multiplied by the difference of the volume parts of the two different domain types across the light path. Under these very strange circumstances, the birefringence of a single $TbPO_4$ domain would come out to be larger (by an unknown factor) than the measured value.

If these two objectives are considered to be unreal, the following consequences must be drawn. Presupposing that the approach of the calculation is applicable to $TbPO_4$ one must realize that the observed oscillations cannot be explained by a monotonic change of the Faraday rotation, of the birefringence or of a combination of both. On the contrary, one has to assume that one or both of these quantities change their strength periodically with temperature. Furthermore it is necessary to assume that this change occurs in such a way that the period of change with temperature is linearly dependent on the field strength. These are really strange conditions. To

tell the truth, we are neither in a position to give the driving mechanism for this behaviour nor to say if such properties are conceivable.

Even the fact that corresponding oscillations were not observed in the measured magnetization, does not absolutely contradict this interpretation, although Faraday rotation and magnetization should to a large extent be proportional. Our knowledge of $TbPO_4$ says that a reduction of the distortion is combined with a reduction of the z component of the magnetic moment of the single Tb^{3+} ion and thus of the magnetization. Consequently the Faraday rotation and birefringence will change in the same direction. Assigning about half the amplitude of the oscillation to a change in birefringence and the other half to a change in Faraday rotation, the amplitude of the oscillations in the magnetization will become so small that they might not be detectable within the experimental error.

4.3. General comments

Let us finally add some words to equations (10) and (11) in general. The results are surprising in different respects. Some of these are the following.

(i) A steady change of Faraday rotation or birefringence or both of them does not necessarily entail a steady variation of the rotation angle.

(ii) Entirely different results are obtained for the two orientations of the optical elements handled above. The reason is that the direction of light modulation (which decides the measured second harmonic of the intensity) is along x, i.e. along a crystallographically distinctive axis, in the first case and at 45° to x in the second.

(iii) The most important finding, however, is that the special angles of α_0 or α_0', at which the plateaux appear or around which the oscillations occur, are multiples of $\pi/2$ or π, respectively. Also the particular values of φ, at which the steps are observed or at which the special values of α_0' are reached, are odd or even multiples, respectively, of $\pi/2$. These values are decided above all by the method of measurement and only in part by the sample to be investigated. As a consequence, measurements on samples of different thicknesses will lead to different results. Thus, the experimental curves do not in every case allow a direct determination of the size of the Faraday rotation and the birefringence. Therefore, care has to be taken in the interpretation of rotation measurements on substances possessing both these effects.

5. Summary

The investigations on $TbPO_4$ have shown that Faraday rotation and magnetization are proportional to each other within the accuracy of measurement for all phases in which the substance has tetragonal symmetry. Due to the comparatively small birefringence in the low-symmetry phases, the deviations from proportionality are modest so that a separate evaluation does not seem reasonable. In one of these phases, however, unusual oscillations were found in the measured rotation as a function of temperature with a period that is almost proportional to the strength of the external field.

A comparison with the results of detailed calculations revealed that the oscillations cannot be explained by a monotonic decrease or increase of Faraday rotation or birefringence. Instead, a temperature-dependent oscillation of one or both of these quantities must be assumed for being able to interpret the observations. This seems a rather strange assumption. But we cannot offer another model that seems more

convincing. Further investigations will be necessary to prove whether the given interpretation is tenable or not.

In addition, the calculations showed that the results of a measurement depend not only on the properties of the substance but also on the experimental arrangement used. This has to be taken into consideration, especially in the investigation of a substance exhibiting a relatively large birefringence in comparison with its Faraday rotation.

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