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Linear magnetic birefringence of the hyperfine enhanced nuclear magnet $HoVO_4$ at low temperature

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Abstract. Measurements of the linear magnetic birefringence of the hyperfine enhanced nuclear magnet $HoVO_4$ in the low-temperature region are reported. It is shown that below 2 K, where only the lowest crystal field level is populated, the temperature dependence of the birefringence is dominated by the paramagnetic polarization of the enhanced nuclear spins. The results are in good agreement with a single-ion model.

1. Introduction

Linear magnetic birefringence has been used widely in the past two decades to study properties of magnetic crystals [1]. It provides a method to detect changes in symmetry due to magnetoelastic distortions of the crystal lattice or due to a change of the electronic wavefunction, which can be caused by magnetic order phenomena or an external magnetic field. It could be shown that in certain cases the change of birefringence shows the same temperature behaviour as the magnetic internal energy [2, 3], which makes possible direct measurements of thermodynamic functions, such as the magnetic specific heat.

Up to now, no experimental or theoretical investigations of the influence of nuclear magnetism on magnetic birefringence have been made. This can be explained by the fact that the phenomena of nuclear magnetism take place at sub-mK temperatures where optical windows to the cryostat can cause severe heating of the sample due to IR radiation. Furthermore, most nuclear magnets are metallic and strongly absorb light in the visible region. Investigations are interesting because the thermodynamic functions in nuclear magnets are not easily accessible due to slow spin-lattice relaxations and difficulties in measuring the temperature of the spin system.

We have chosen holmium vanadate ($HoVO_4$) as a test system for birefringence measurements of nuclear magnets. $HoVO_4$ is an insulating compound which shows 'enhanced nuclear magnetism'. The enhancement of the ^{165}Ho nuclear spins ($I = \frac{7}{2}$) results from a coupling of the electronic magnetic moment to the nuclear spins due to a strong hyperfine interaction and the singlet ground-state of the electronic system. The properties of the electronic and the nuclear system of $HoVO_4$ are well known [4–8]. Below $T_N = 4.5\,mK$ the nuclear spins undergo an antiferromagnetic phase transition, which has been studied with different techniques [9–12].

Here we present our measurements on the temperature dependence of the birefringence of $HoVO_4$ at low temperatures. In section 3 we show that at low temperatures the

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birefringence is driven by the field-induced nuclear polarization which couples via a hyperfine interaction to the 4f electrons of the Ho^{3+} ion. For the nuclear spin system in the temperature range around 1 K paramagnetic behaviour is expected. This is indeed identified in our experimental data presented and discussed in sections 4 and 5.

2. Experimental setup

We measured the birefringence using an arrangement similar to that described by Sénarmont [13] with an additional phase-modulation technique. Due to the geometry of the cryostat and magnet the laser beam ($\lambda = 632.8$ nm) had to be deflected to the sample by four gold-coated mirrors; another set of four mirrors was needed to extract the beam from the experimental space for polarization analysis. Constant phaseshifts caused by the mirrors do not disturb the measurement as we are interested only in changes of birefringence. The sample chamber had to be shielded against the thermal room-temperature radiation entering through the optical access tubes. This was achieved by mounting standard IR filters in a stress-free manner at liquid helium temperature. Neither the signal quality nor its long time-stability was affected by this. The power of the laser (4 mW) was reduced to 2.5% in order to minimize beam heating of the sample. Measurements at different attenuations of the laser beam showed no evidence of beam heating above 1 K, whereas at lower temperatures beam heating is the most probable explanation for deviations from paramagnetic behaviour.

Our single-crystal sample had an almost cubic shape and a length of 0.78 mm along the [001] direction. Parallel (001) planes were prepared by grinding to an accuracy of 0.2°. The sample was glued to the sample holder only at one side in order to avoid strains. We estimate the error in sample orientation with respect to the laser beam to be below 1°. A mask (0.5 mm) in front of the sample was used to select a small region of the crystal.

The sample was mounted to the cold finger of a ^{3}He cryostat in the centre of a 9 T superconducting solenoid. The temperature of the sample could not be monitored directly; instead, a Ge resistor and a 100 Ω Speer resistor mounted close to the sample holder were used to determine the temperature. The field dependence of the Speer resistor was calibrated using the ^{3}He vapour pressure as reference.

3. Theory

3.1. Zeeman effect and enhanced nuclear magnetism

In $HoVO_4$ the magnetic ion Ho^{+3}, $4f^{10}$, $^5I_8$, occupies a site of tetragonal symmetry. Neglecting interactions a single-ion Hamiltonian can be written as

$$\mathcal{H} = \mathcal{H}_{CF} + g_J \mu_B \boldsymbol{J} \cdot \boldsymbol{B} - g_N \mu_N \boldsymbol{I} \cdot \boldsymbol{B} + A_J \boldsymbol{J} \cdot \boldsymbol{I} + \mathcal{H}_Q. \tag{1}$$

The terms in (1) represent the interaction with the crystal field, the electronic and nuclear Zeeman interaction, the magnetic hyperfine interaction and the electric quadrupole hyperfine interaction. To describe the influence of magnetic fields or temperature changes on birefringence one has to solve $\mathcal{H}$ to obtain the electronic wavefunctions of the thermally occupied states as magnetic birefringence arises through virtual dipole transitions to excited states.

The ground manifold $^5I_8$ is split by $\mathcal{H}_{CF}$ into four doublets and nine singlets in such a way that the ground state is a singlet which is almost pure $|J_z = 0\rangle$ [8]. Following

Battison and co-workers [4], as well as the ground-state singlet, a doublet at $\epsilon = 29.9$ K (predominantly $|J_z = \pm 1\rangle$) and a singlet at $\epsilon' = 66.9$ K (predominantly $|J_z = 2^s\rangle$) have to be considered to describe the Zeeman interaction of the low-lying states. With the magnetic field lying in the tetragonal plane the electronic Zeeman interaction is $\frac{1}{2}B(J_+e^{i\varphi} + J_-e^{-i\varphi})$, where φ is the angle of the field with respect to [100]. To solve the Hamiltonian we neglect the quadrupolar interaction and the components of the nuclear spins perpendicular to the magnetic field. This approximation used by Bleaney [14] to explain the enhanced nuclear spins leads to a separation of the nuclear and electronic spin operators. Within these approximations we can write (1) in matrix form:

$$\begin{array}{c} \\ \langle 0| \\ \langle 1| \\ \langle -1| \\ \langle 2^s| \end{array} \begin{array}{c} \begin{array}{cccc} |0\rangle & |1\rangle & |-1\rangle & |2^s\rangle \end{array} \\ \begin{pmatrix} h_n & h_1 e^{-i\varphi} & h_1 e^{i\varphi} & 0 \\ h_1 e^{i\varphi} & \epsilon + h_n & 0 & h_2 e^{-i\varphi} \\ h_1 e^{-i\varphi} & 0 & \epsilon + h_n & h_2 e^{i\varphi} \\ 0 & h_2 e^{i\varphi} & h_2 e^{-i\varphi} & \epsilon' + h_n \end{pmatrix} \end{array} \tag{2}$$

where

$$h_1 = \tfrac{1}{2}(g_J \mu_B B + A_J \langle I_\varphi \rangle)\alpha_1 \qquad \alpha_1 = \langle 8, 0|J_\pm|8, \mp 1\rangle$$
$$h_2 = \tfrac{1}{2}(g_J \mu_B B + A_J \langle I_\varphi \rangle)\alpha_2 \qquad \alpha_2 = \langle 8, 2|J_\pm|8, \pm 1\rangle$$
$$h_n = -g_N \mu_N I.$$

Here I_φ represents the component of the nuclear spin along the field. The ground-state energies and wavefunctions for fields along [100] and [110] are easy to calculate and are shown in table 1. To get compact analytic expressions we use the approximation $\epsilon' = 2\epsilon$ and $\alpha_1 = \alpha_2$ as suggested by Bleaney [6].

Table 1. Ground-state energies and electronic wavefunctions for the different field directions.

	Ground-state energy	Electronic wavefunction
$B\|\|$ [100]	$E_{100} = \epsilon - W$ with $W = (\epsilon^2 + 2h^2)^{1/2}$, $h^2 = h_1^2 + h_2^2$	$\|\psi_{100}\rangle = [h^2/W(W-\epsilon)]\|0\rangle - (h/W)\|1^s\rangle + [h^2/W(W+\epsilon)]\|2^s\rangle$
$B\|\|$ [110]	$E_{110} = \frac{1}{2}\epsilon - (\frac{1}{4}\epsilon^2 + 2h_1^2)^{1/2}$	$\|\psi_{110}\rangle = \cos\theta\|0\rangle - e^{i\pi/4}\sin\theta\|-1'\rangle$ with $\|\pm 1'\rangle = (1/\sqrt{2})(\|1\rangle \pm i\|-1\rangle)$ and $\tan 2\theta = -2\sqrt{2}h_1/\epsilon$

Note that now the thermal average of the nuclear polarization $\langle I_\varphi \rangle$ enters the ground-state energy as well as the electronic wavefunction through h_1 and h_2. Within this approximation, therefore, a change in $\langle I_\varphi \rangle$ will be observable in the birefringence just like a change in the externally applied field.

A low-field expansion of the ground-state energies yields the Van Vleck paramagnetism of the electronic system and the enhanced nuclear Zeeman Hamiltonian of the nuclear spins as has been described in the literature [5, 6, 8]. The polarization of the nuclear spins in this field regime at temperatures $T \gg T_N$ will show paramagnetic behaviour:

$$\langle I_\varphi \rangle = C_I \frac{B}{T} \tag{3}$$

where C_I is the hyperfine enhanced Curie constant. For high fields the enhancement of the nuclear spins vanishes. The energy splitting ΔE_n is then determined by the hyperfine interaction and the electronic saturation magnetization.

3.2. Effects on birefringence

In $HoVO_4$ magnetic birefringence at $T \gg T_N$ can be explained by a single-ion model. Two possible cases, virtual excitations from a low-lying doublet to higher singlets, where the doublet is split by a magnetic field, and excitations from a low-lying singlet to split doublets, are discussed by Becker and Gehring [15]. They find two contributions to birefringence

$$\Delta n_{\text{doub}} = (p_{+1} - p_{-1})A(\omega)$$
$$\Delta n_{\text{sing}} = p_s e_{ij} B(\omega) \tag{4}$$

where p_{+1}, p_{-1} and p_s are the populations of the split ground doublet and the ground singlet, e_{ij} is a magnetoelastic strain of appropriate symmetry which splits excited doublets linearly, and $A(\omega)$ and $B(\omega)$ are frequency-dependent factors which can be adjusted to experimental findings. For $HoVO_4$ the magnetic birefringence for temperatures where only the ground state is populated and for the two field directions is then calculated as

$$\Delta n_{100} = A_{100}\frac{h^2}{W^2} + B_{100,0}\frac{h^6}{W^4(W-\epsilon)^2} \tag{5}$$

and

$$\Delta n_{110} = A_{110}\sin^2\theta + B_{110}\sin^2\theta\cos^2\theta \tag{6}$$

where h, W and θ are defined in table 1. These equations describe the birefringences associated with a change of the applied field, which in our case is the sum of the external and the small hyperfine fields. The effect of the polarized Ho nuclei on the birefringence is obtained by series expansion of (5) and (6) respectively. This yields for both orientations to lowest order

$$\Delta n = \Delta n_0 + \frac{D}{T} + \ldots \tag{7}$$

because $\langle I_\varphi \rangle$ varies linearly with T^{-1}. The slope $D = D(B)$ is a function of the field B and depends on the matrix elements α_i, the gap ϵ, the hyperfine coupling A_J which are known from the literature [4, 5], the constants $A(\omega)$ and $B(\omega)$, which can be determined experimentally by field sweeps, and on the Curie constant in (3).

4. Results

We performed field sweeps with fields along [100] and [110]. The parameters $A(\omega)$ and $B(\omega)$ were obtained consistently by fitting the experimental data to (5) and (6). The results of Becker and Gehring [15] could be reproduced with these measurements.

Temperature sweeps for the two different field directions and different fields are shown in figure 1. The measured birefringence is shown as a function of reciprocal temperature. All experimental data show a proportionality to T^{-1} in a temperature range between 1–2 K. In this linear region straight lines were fitted to the data. They are shown in figure 1 as broken lines. Above 2 K due to thermal population of higher crystal field levels an additional birefringence signal appears. In figure 1 this is seen as a steep decrease of the signal at temperatures above 2 K. Below 1 K the sample temperature differs from the

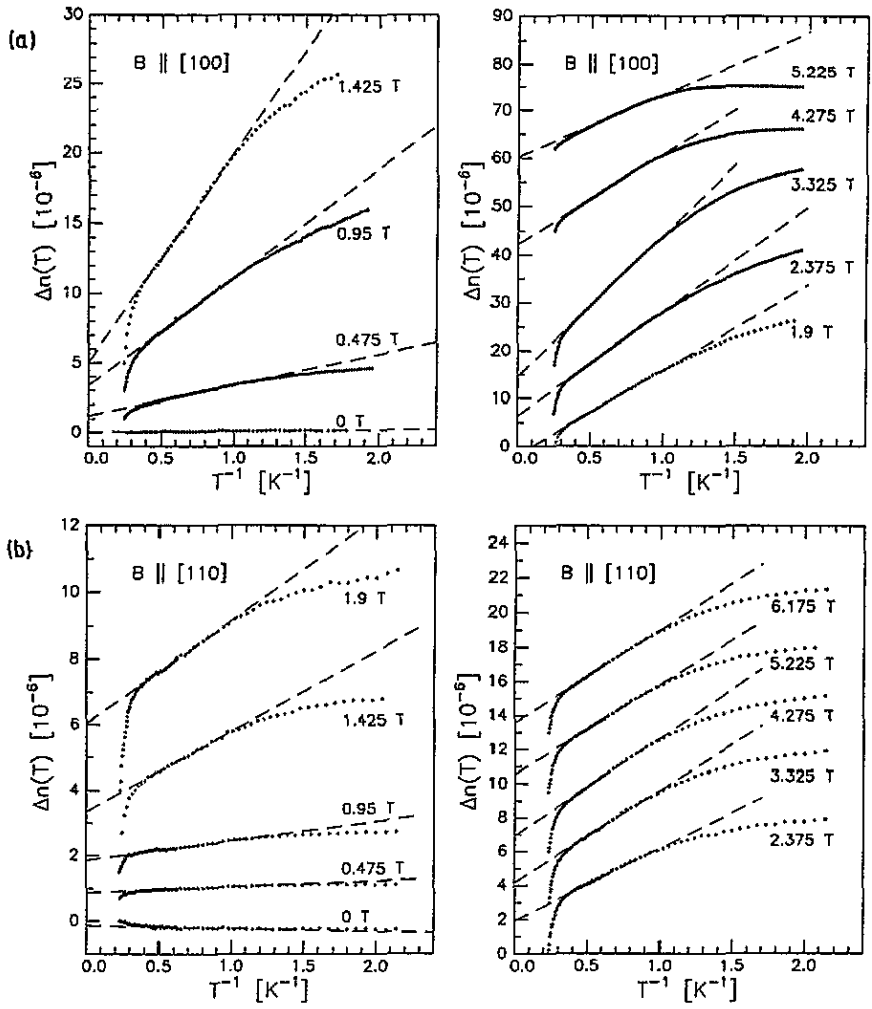

Figure 1. Temperature dependence of the birefringence for different fields along (*a*) [100] and (*b*) [110]. The change of the birefringence is shown as a function of the reciprocal temperature with an arbitrary offset for each curve.

measured temperature of the sample holder, which is explained through beam heating of the sample (see section 2). We estimate the base sample temperature to be 0.6 K, whereas the sample holder could be cooled to 0.4 K in our cryostat.

The change of birefringence in the temperature sweeps is three magnitudes smaller than observed in the field sweeps. This justifies the linear approximations to the birefringence made in the previous section.

We extracted the slopes of the linear region between 1–2 K. They are shown in figure 2. The error bars result from the uncertainty in the thermometry and the internal field due to demagnetizing effects. The full curves are theoretical curves obtained by combining (5) and (6), respectively, with the appropriate wavefunctions and the measured $A(\omega)$ and $B(\omega)$ values. Here the nuclear Curie constant is the only free parameter; it was adjusted to fit the experimental data. From temperature sweeps with field applied along the [100] direction we find the value $C_I = (0.20 \pm 0.01) K/T$, whereas a high-temperature expansion

of the Brillouin function with the 'enhancement factor' $K = 170$ [5] gives the value $C_I = K[I(I+1)/3I]g_n\mu_n/k_B = 0.36K/T$.

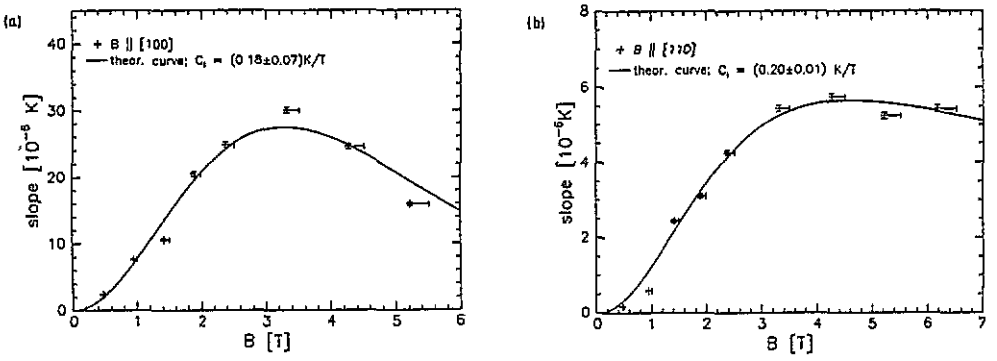

Figure 2. Measured slopes of the T^{-1}-dependent birefringence signal between 1 K and 2 K and theoretical curve calculated following (7) for fields along (*a*) [100] and (*b*) [110].

5. Discussion

In the foregoing we have shown that in $HoVO_4$ the birefringence at temperatures around 1 K is induced by the polarized Ho nuclei. Using a single-ion model and known values for the hyperfine interaction we were able to measure the Curie constant consistently for two different crystal orientations. The deviation by roughly a factor of two compared to values in the literature might be due to impurities, which can alter the observed enhancement factor in our sample. Other effects, like quadrupolar interactions or a coupling to magnetoelastic strains [16], are not expected to be important at these temperatures. They may be more important at higher nuclear polarizations.

Experimental difficulties are mainly due to problems of sample thermalization at temperatures below 1 K. A major source of external heat input to the sample was room temperature radiation entering our sample chamber. This could be removed by using IR filters at low temperatures. The other problem is the heating of the sample due to the laser beam. In our setup, the detection of the signal offered no serious problem, thus further attenuation appears feasible. Another possibility would be the choice of a wavelength which minimizes absorption. This, however, was not possible with our present light source.

To conclude, we have demonstrated that magnetic birefringence data in $HoVO_4$ can be obtained below 1 K. The present experiments were performed mainly to study the paramagnetic regime of the Ho nuclei. It will be interesting to extend the temperature range of these experiments down to 4.5 mK where the nuclei order. Extrapolating our data to these temperatures, we expect a birefringence signal of 2×10^{-6} which can be detected easily. Close to T_N we expect the single-ion model to break down, giving a birefringence signal which is proportional to the internal energy of the nuclear system. This is of particular interest, as it will give a measure for this thermodynamic function in an adiabatically cooled system obtained by a method which does not rely on calorimetry.

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