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Magneto-elastic properties of $Tb_3Ga_5O_{12}$ (TGG)

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Abstract. The temperature dependence of the elastic constants in $Tb_3Ga_5O_{12}$ was measured and analysed with a simple crystal field model. The magneto-elastic coupling constants have been deduced from this experiment. The coupling constant $g_{\Gamma 5}$, related to the c_{44} mode, is anomalously large. These coupling constants are important for the interpretation of the phonon Hall effect.

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Rapid Note

Recently the so-called phonon Hall effect was measured in the garnet substance $Tb_3Ga_5O_{12}$ (TGG) [1]. Applying a magnetic field perpendicular to the heat current flowing through this insulating paramagnetic crystal a temperature difference between the edges of the sample in the third direction is observed. Theories trying to explain this effect [2,3] are taking a magneto-elastic Hamiltonian which is used for spin-lattice relaxation studies and other magneto-elastic effects. Therefore a determination of the corresponding magneto-elastic coupling constants is important.

The temperature dependence of the elastic constants for crystal electric field (CEF) systems like TGG depends strongly on the magneto-elastic coupling arising from the strain modulation of the CEF. This coupling can be described with the magneto-elastic Hamiltonian

$$H_{me} = \Sigma_{\Gamma,i} g_\Gamma \varepsilon_\Gamma O_{\Gamma,i} \qquad (1)$$

where ε_Γ are the symmetry components of the strain tensor and O_Γ the quadrupolar operators of symmetry Γ respectively. The magneto-elastic coupling constant g_Γ can conveniently be determined with the temperature dependence of the elastic constants [4].

The temperature dependence of the sound velocities v was measured by a homemade ultrasonic apparatus based on a phase comparison method. The elastic constants were calculated with $c = \rho v^2$ with the mass density $\rho = 7.22$ g/cm^3. The ultrasonic waves were generated and detected by a pair of $LiNbO_3$ transducers bonded on the parallel surfaces of the sample. The low temperature measurements were performed using a ^{3}He-evaporation refrigerator.

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In Figure 1 we show the temperature dependence of the elastic constants c_{11}, $(c_{11}-c_{12})/2$ and c_{44}. All 3 modes exhibit pronounced anomalies below 150 K with strong minima around 30–50 K and much smaller anomalies below 4 K (as shown in the inset of the figure). As an example the c_{44}-mode exhibits the largest softening of more than 5% from 300 K to 30 K. The small minimum at 2 K amounts to 0.1% only. The bulk modulus $c_B = (c_{11} + 2c_{12})/3$ on the other hand is only weakly temperature dependent in the whole temperature region. Its temperature variation for $T < 150$ K is less than 0.3%. This points to magneto-elastic effects for the elastic constants due to the strain-quadrupolar coupling of the Tb^{3*}-ion as described with equation (1).

$Tb_3Ga_5O_{12}$(TGG) has the cubic garnet structure (space group O_h^{10}). The rare earth ion is on a position with D_2 point symmetry. The Tb^{3+} $(4f^8)$ - ion has $S = 3, L = 3$ and $J = L+S = 6$. Due to the low site symmetry the crystal electric field (CEF) levels are split into $2J+1 = 13$ singlets. No magnetic order occurs down to 2 K. TGG represents a frustrated spin system, like other magnetic substances with the garnet structure.

The crystal field parameters for TGG have been determined [5] and the crystal field levels consist of 2 low lying levels, approximately 3–4 K apart and of 4 excited levels around 60 K. All other levels are higher than 150 K and are neglected. This level schema allows a simplified calculation of the strain susceptibilities for the temperature dependence of the symmetry elastic constants. Returning to Figure 1 qualitatively the small anomalies below 4 K are due to the strain coupling to the low lying doublet and the large anomalies around 30 K are due to the higher lying levels around 60 K. We can use a simple model by relating the CEF level structure of TGG with the corresponding cubic CEF manifold for $J = 6$ [6]. In this model the low

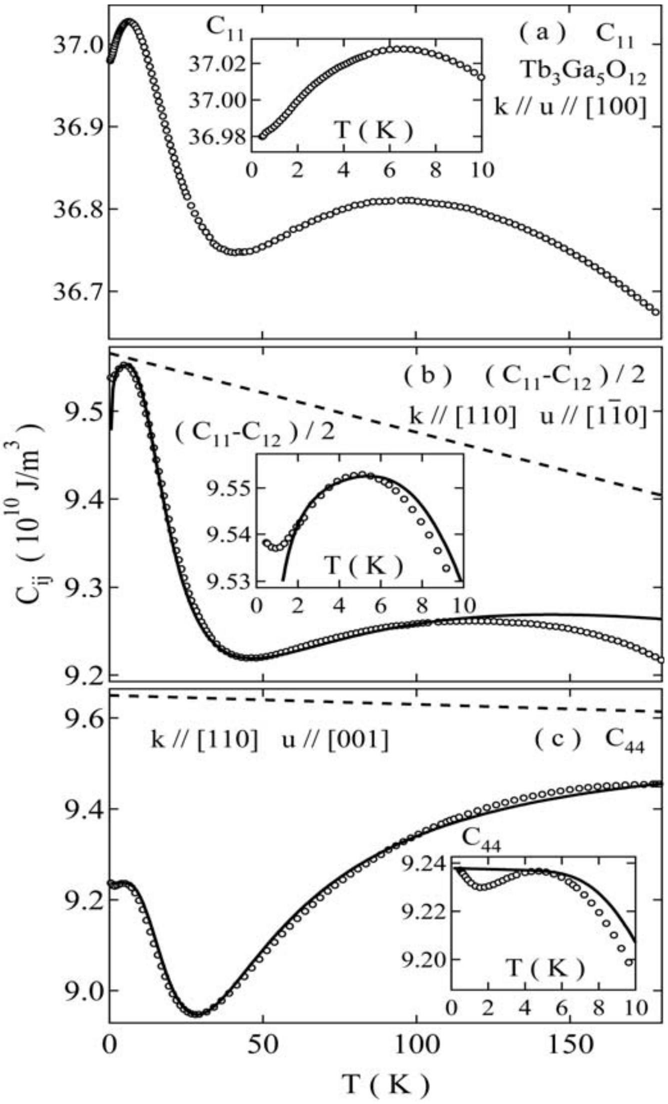

Fig. 1. Temperature dependence of elastic constants (a) c_{11}; (b) $1/2(c_{11} - c_{12})$; (*c*) c_{44}. Full lines are the fits with equations (2), (3); dotted lines are background elastic constants c_o.

lying doublet corresponds to a degenerate $\Gamma_3(E)$ doublet in the cubic case and the 4 singlets to a degenerate $\Gamma_5(T_2)$ triplet and $a\Gamma_2(A_2)$ singlet. From the tables of reference [6] we get with $x = 0.8525$ and $W = -24.5$ a level scheme of $\Gamma_3(0\text{ K}) - \Gamma_5^2(57.3\text{ K}) - \Gamma_2(190\text{ K})$ and all other levels are at much higher energies. By reduction of symmetry from cubic to orthorhombic we get a lifting of degeneracy of the energy levels $\Gamma_3(E) \rightarrow A + B_1$ and $\Gamma_5(T_2) \rightarrow B_1 + B_2 + B_3$ and $\Gamma_2(A_2) \rightarrow A$ where A, B_1, B_2, B_3 denote the symmetry labels of the singlet states in the orthorhombic symmetry. We use the cubic CEF model only to calculate the matrixelements $\langle \Gamma_5/O_\Gamma/\Gamma_5 \rangle$ for $O_{\Gamma 3} = O_o^2 = 3J_z^2 - J(J+1)$ and for $O_{\Gamma 5} = O_{xy} = 1/2(J_xJ_y + J_yJ_x)$ and analogously for the Γ_3 states. We get $\langle \Gamma_5^2/O_{xy}/\Gamma_5^2 \rangle = 6$ and $\langle \Gamma_5^2/O_2^o/\Gamma_5^2 \rangle = -32$ and $\langle \Gamma_3/O_2^o/\Gamma_3 \rangle = \pm 36$. Other matrixelements are gained from the fit to the experiment, see below.

Elastic constants in paramagnetic crystals, which show crystal field levels from the magnetic ions, exhibit anomalies which can be calculated from equation (1) [4]. The change in the elastic constants is

$$c_\Gamma - c_\Gamma^o = -g_\Gamma^2 n \chi_\Gamma / (1 - g'_\Gamma \chi_\Gamma) \qquad (2)$$

with g_Γ the magneto-elastic coupling constant. n denotes the number of magnetic ions per cm^3, in our case , the Tb^{3+}-ions, using the mass density from above, with $n = 1.28 \times 10^{22}/cm^3$. χ_Γ is the strain susceptibility and it is the sum of χ_{str}^C and χ_{str}^{VV} which are, in analogy to the magnetic susceptibility, the Curie type and Van Vleck type strain susceptibilities, $\chi_\Gamma = \chi_{str}^C + \chi_{str}^{VV}$. They are due to the diagonal quadrupolar matrixelements (Curie type χ_{str}^C) and off-diagonal matrixelements (Van Vleck type χ_{str}^{VV}). g'_Γ is the quadrupolar inter-site coupling constant. Many CEF systems have been observed and analysed with this formalism, for a review see e.g. [4].

From Figure 1 we can conclude that the large minima around 30–40 K can be described by a Curie type strain susceptibility of the triplet state around 60 K and the small minima around 1–2 K by a Curie strain susceptibility of the low lying doublet. Furthermore only the c_{44} mode has a much reduced elastic constant value at low temperature which indicates a contribution of a strong Van Vleck type strain susceptibility in this case.

The explicit expressions for the strain susceptibilities are for our model:
For $(c_{11} - c_{12})/2$

$$\chi_{str}^C = [\alpha^2 \langle \Gamma_3/O_2^o/\Gamma_3 \rangle^2 \exp(-\Delta_d/T) + \langle \Gamma_5/O_2^o/\Gamma_5 \rangle^2 \exp(-\Delta_t/T)]/(TZ)$$

For c_{44}

$$\chi_{str}^C = \langle \Gamma_5/O_{xy}/\Gamma_5 \rangle^2 \exp(-\Delta_t/T)/(TZ).$$
$$\chi_{str}^{VV} = \beta^2 \langle \Gamma_3/O_\Gamma/\Gamma_5 \rangle^2/(\Delta_t - \Delta_d) \times (1 - \exp(-\Delta_t/T))/Z. \qquad (3)$$

Here Z is the partition function for the corresponding levels and Δ_d, Δ_t the energy splitting for the doublet and the doublet-triplet separation. with the matrixelements given above. The factors α and β are fitting parameters for the matrixelements. Especially the lower doublet has strongly diminished matrixelements (with fitting parameter α) as can be seen from the experimental results.

As pointed out above the 3 symmetry elastic constants have anomalies which arise mainly from the diagonal quadrupolar matrixelements (Curie terms). Only for the c_{44} with a much reduced elastic constant at low temperature the Van Vleck contribution is sizeable.

The temperature dependence of the 3 elastic constants c_{11}, $(c_{11} - c_{12})/2$ and c_{44} were fitted with the expressions of equations (2), (3). The fit parameters Δ_t, c_Γ^o, α, β and g_Γ, g' are given in Table 1. The choice of the background elastic constants c_Γ^o is indicated in the figure by dotted lines. The fit is very good for the temperature region 4–150 K for all the elastic modes, especially the symmetry modes $(c_{11} - c_{12})/2$ and c_{44}. The low temperature anomaly due to the two A states is qualitatively described only for

Table 1. Fit parameters for the elastic constants.

	c_{11}	$(c_{11} - c_{12})/2$	c_{44}
Quadrupolar operator O_Γ	O_o^2	O_o^2	O_{xy}
Δ_t (K)	57	57	57
$c_\Gamma^o (10^{10}\,J/m^3)$ ($T = 0\,K$)	37.3	9.57	9.63
Matrixelement parameters α^2		0.0015	
β^2			0.45
$/g_\Gamma/$ (K)		62	601
g' (mK)		−15	6.5

the $(c_{11} - c_{12})/2$ mode. For the c_{44} mode the Curie term for the lower doublet is negligible.

In order to estimate the magneto-elastic coupling constants g_Γ we take the quadrupolar matrixelements from our simple $\Gamma_3 - \Gamma_5 - \Gamma_2$ model listed above. Including these with the fitting parameters α, β gives the coupling constants listed in Table 1. Note that the matrixelements for our cubic model, together with the fitting parameters, should give a good order of magnitude estimate. c_{44} has by far the largest coupling constant, an order of magnitude larger than the Γ_3 mode $1/2(c_{11} - c_{12})$.

The intersite coupling constants g'_Γ has a different sign for the c_{44} and $1/2(c_{11} - c_{12})$ modes. It has a ferroquadrupolar interaction for the c_{44}-mode and an antiferroqudrupolar interaction for the $1/2(c_{11} - c_{12})$-mode.

For the phonon Hall effect one uses a spin-phonon interaction term [2,3] of the form $H_{sph} = G\Sigma_i\ \sigma_i\ [\mathbf{u}_i \times \mathbf{p}_i]$ with the vectors of displacement $\mathbf{u}$, momentum $\mathbf{p}$ and pseudospin σ for the lowest doublet. This type of coupling was derived [7] from equation (1) with G proportional to $g_{\Gamma 5}^2$ for the case of $CeAl_2$ which exhibits also strong CEF effects [4]. Our analysis above gives for $g_{\Gamma 5}$ also a very large value for TGG. This $g_{\Gamma 5}(c_{44})$ is responsible for the phonon Hall effect.

A full account of this work, together with ESR results of the CEF-levels, magnetic field dependence of the various elastic constants, magnetic susceptibility measurements and a more detailed discussion will be given in the future. The single crystal of TGG was kindly given to us by P. Wyder.

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