# MAGNETIC EXCITATIONS IN TbP *

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The magnetic $\Gamma_1 - \Gamma_4$ exciton of the singlet ground state system TbP has been studied by inelastic neutron scattering above the antiferromagnetic ordering temperature. It showed considerable dispersion and a pronounced splitting in the [001] and the [110] directions which increased as the transition temperature was approached in accordance with the predictions of the RPA-theory

The ground multiplets of the rare-earth ions in solids are split by the crystal field. Magnetic dipole transitions of finite energy transfer are possible between the crystal-field states. In the presence of an interionic magnetic coupling these excitations will propagate through the lattice and will form bands. Systems with nonmagnetic singlet ground states have attracted special interest for the magnetic ordering can only be achieved by a polarisation of the ground-state wave function. In the paramagnetic phase one expects a soft mode behavior of the magnetic excitations at the ordering wave vector [1]. We will show that TbP is an almost ideal candidate for the study of the dynamics of these systems.

TbP crystallizes in the NaCl-structure ($a = 5.68$ Å). The $J = 6$ ground multiplet of the $Tb^{3+}$-ions is split by the crystal field into a singlet $\Gamma_1$ ground state and triplets $\Gamma_4$ and $\Gamma_5$ at 1.63 and 3.47 meV followed by three further levels above 10 meV [2,3]. At $T_N$ = 7.3 K the system orders antiferromagnetically with an ordering wave vector $\boldsymbol{q}_0 = (\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2})$ which is the L-point of the Brillouin zone [4]

The inelastic neutron scattering experiments on the low-lying magnetic excitations in the paramagnetic regime were carried out on the triple axis spectrometers TAS1 and TAS6 at the cold source of the reactor at Risø. Either the incident or the outgoing neutron energy was fixed at 5 meV. The data were collected in the constant-$\boldsymbol{Q}$-mode of operation. Energy transfers up to about 3 meV could be reached. The TbP sample contained a single crystalline grain of about 0.5 cm$^3$. The crystal was oriented with the [110] direction perpendicular to the scattering plane.

The cross-section for inelastic magnetic neutron scattering is a direct measure of the imaginary part of the dynamic magnetic dipole susceptibility $\chi(\boldsymbol{q}, \omega, T)$. The energies and the mode strengths of the magnetic excitations are determined by the poles and the corresponding residues of the susceptibility [1]. Within the RPA-theory, $\chi$ is related to the single ion crystal field susceptibility $u(\omega, T)$

$$u(\omega, T) = \sum_{i \neq j} \frac{|M_{ij}|^2 \Delta_{ij}(n_i - n_j)}{\Delta_{ij}^2 - \omega^2} + \frac{\delta_{\omega,0}}{k_B T} \sum_i |M_{ii}|^2 n_i$$

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via

$$\chi^{\alpha\beta}(q, \omega, T) = \frac{g^2 \mu_B^2 u(\omega, T)}{1 - J^{\alpha\beta}(q) u(\omega, T)} \quad \text{with } \alpha, \beta = x, y, z,$$

where $J(q)$ is the Fourier transform of the exchange coupling, $\Delta_{ij}$ and $|M_{ij}|^2$ are the energy splittings and the magnetic dipole matrix elements between the crystal field states $i$ and $j$ which are thermally populated according to the Boltzmann factors $n_i$ resp. $n_j$ [1]. In a cubic system $u(\omega, T)$ is a scalar but $J^{\alpha\beta}$ and $\chi^{\alpha\beta}$ are in general tensor quantities.

At high temperatures (>100 K), the neutron experiment showed scattered neutron groups around 1.6 meV independent of the momentum transfer. According to the crystal-field level scheme this signal is identified as a superposition of the $\Gamma_1 - \Gamma_4$ and the $\Gamma_4 - \Gamma_5$ crystal field transitions. At lower temperatures the dispersion develops. The energies of the excitations propagating along the main symmetry directions at 7.9 K are shown in fig. 1. The assignment of the different branches was achieved by a systematic study of the dependence of the energies and intensities of the scattered neutron groups on the temperature and the momentum transfer. The weak signals around 1.6 meV (triangles in fig. 1) represent the $\Gamma_4 - \Gamma_5$ transition which is predicted to remain dispersion-free down to $T_N$ by RPA-theory apart from those regions in reciprocal space where it anticrosses the $\Gamma_1 - \Gamma_4$ exciton. The two other branches (solid circles) belong to the $\Gamma_1 - \Gamma_4$ exciton. They show considerable dispersion and a splitting for finite reduced wave vectors $q$ in the [001] and the [110] direction. The two branches are characterized by their polarisation with respect to $q$. The band width of the $\Gamma_1 - \Gamma_4$ exciton grows when approaching $T_N$ from above, at the L-point a soft mode behaviour with a concomitant central peak was observed. The temperature dependence of the soft mode is shown and compared to the RPA result in fig. 2. The relevant parameters of the RPA calculation are $\Delta(\Gamma_1 - \Gamma_4) = 1.60$ meV and $J(L) = 0.061$ meV. These parameters yield a divergence of the static susceptibility at 5.5 K which is 1.8 K lower than the experimental value for $T_N$. This result and the steplike jump of the excitation energy to 2.0 meV are strong indications for the phase transition being discontinuous. The splitting of the $\Gamma_1 - \Gamma_4$ exciton is surprising. Under the usual assumption of an isotropic coupling the degeneracies of the single ion crystal field

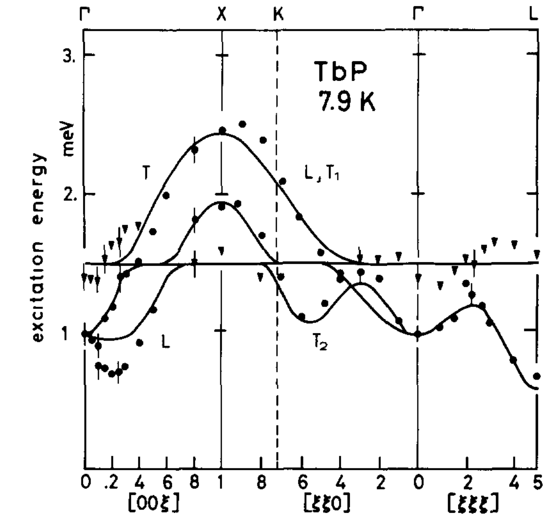


Fig. 1. The dispersion relation of the low energy excitations in the three main symmetry directions at 7.9 K. The two branches of the $\Gamma_1 - \Gamma_4$ exciton are given by solid dots and are characterized by their longitudinal ($L$) or their transverse ($T$) polarisation The $\Gamma_4 - \Gamma_5$ exciton is given by triangles. The solid line is the best fit to the RPA-theory using a phenomenological anisotropic coupling

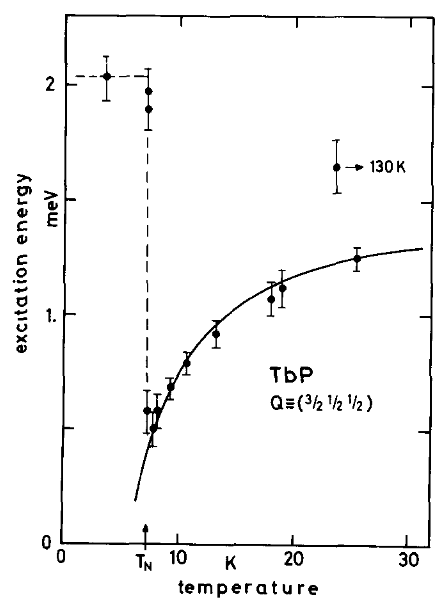


Fig. 2. The temperature dependence of the $\Gamma_1 - \Gamma_4$ excitation energy at the L-point. The solid line is the result of the RPA-theory. The dashed line is a guide for the eye.

transitions are carried over on the propagating excitons. In that case the $\Gamma_1 - \Gamma_4$ exciton should be threefold degenerate for all $\boldsymbol{q}$. The splitting is a proof that $J(\boldsymbol{q})$ is anisotropic. In the analysis of the present results we considered all the symmetry allowed coupling tensors up to the fourth nearest Tb neighbours. In order to reproduce the observed degeneracies it was necessary to demand that all the tensors were diagonal. The number of free parameters of this model is then two per neighbour shell. The best fit to the experimental data is shown as a solid line in fig. 1. At present we are not able to specify a Hamiltonian for this tetragonal type of anisotropic coupling. In particular this special interaction is different from the classical dipole–dipole coupling. Kaplan and Lyons [5] showed in studying the exchange due to conduction electrons that the dipolar terms are by no means sufficient for a complete description. Higher order terms could be responsible for the cancellation of the off-diagonal elements of the coupling tensors.

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## References

[1] W.J.L. Buyers, AIP Conf. Proc. 24 (1975) 27.
[2] R.J. Birgeneau, E Bucher, J.P. Maita, L. Passell and K.C Turberfield, Phys. Rev. B8 (1973) 5345.
[3] E Bucher, J.P. Maita, G.W. Hull, Jr., L.D. Longinotti, B. Lüthi and P.S. Wang, Z. Phys. B25 (1976) 41.
[4] H.R. Child, M.K. Wilkinson, J.W. Cable, W.C. Koehler and E.O. Wollan, Phys. Rev. 131 (1963) 922.
[5] T.A. Kaplan and D.H. Lyons, Phys. Rev 129 (1963) 2072.