acta phys. slov. 39 (1989), No. 4

MAGNETIZATION BEHAVIOUR OF THE $Tb(Cu_{0.7}Ni_{0.3})_2$ SINGLE CRYSTAL [1)]

ПОВЕДЕНИЕ МАГНЕТИЗАЦИИ ПРОСТОГО КРИСТАЛЛА $Tb(Cu_{0.7}Ni_{0.3})_2$

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$Tb(Cu_{0.7}Ni_{0.3})_2$ belongs to the heavy $RE(Cu_{1-x}, Ni_x)_2$ ($x \leq 0.3$) series of intermetallic compounds which all crystallize in the $CeCu_2$ orthorhombic crystallographic structure with the space group Imma (D_{2h}^{28}) [1]. This compound exhibits the ferromagnetic ordering with the Curie temperature $T_c \approx 85$ K [1, 2]. The neutron diffraction experiments confirmed this magnetic structure and showed that only the Tb ion had a magnetic moment orientated in an $\boldsymbol{a}-\boldsymbol{c}$ plane without any contribution from the Ni ions [3]. The measurements of single crystal magnetization in low [2] and high [4] magnetic fields yielded strong anisotropy along the principal crystallographic axes. The field dependence of magnetization in the $\boldsymbol{b}$ direction increases linearly up to 18 T, where there appears a sudden jump and the magnetization reaches its saturated value (see Fig. 1). This interesting behaviour was described in terms of the phenomenological magnetocrystalline anisotropy [4]. Our work aims at explaining these facts from the nature of the crystal electric field (CEF) in this compound. We can show that the crossing of the lowest energy levels of Tb in the magnetic field leads to the step-like appearance of the low temperature magnetization curves in the $\boldsymbol{b}$ direction.

We have added a Zeeman term describing the interaction of the Tb magnetic moment with the external magnetic field $\boldsymbol{H}$ and the exchange field $\boldsymbol{H}_m = \lambda \boldsymbol{M}$ in the molecular field approximation to the full Hamiltonian, which can be written in the form:

$$\hat{H} = \hat{H}_{CEF} - g\mu_B(\boldsymbol{H}_m + \boldsymbol{H})\cdot\hat{\boldsymbol{J}}, \qquad (1)$$

where

$$\hat{H}_{CEF} = V_2^0\hat{O}_2^0(\hat{J}) + V_2^2\hat{O}_2^2(\hat{J}) + V_4^0\hat{O}_4^0(\hat{J}) + V_4^2\hat{O}_4^2(\hat{J}) + V_4^4\hat{O}_4^4(\hat{J}) + $$
$$+ V_6^0\hat{O}_6^0(\hat{J}) + V_6^2\hat{O}_6^2(\hat{J}) + V_6^4\hat{O}_6^4(\hat{J}) + V_6^6\hat{O}_6^6(\hat{J}) \qquad (2)$$

is the CEF Hamiltonian of the orthorhombic symmetry (V_n^m are the CEF parameters and $\hat{O}_n^m(\hat{J})$ are the Stevens equivalent operators), g is the Landé factor ($g = 1.5$), μ_B is the Bohr magneton and $\hat{\boldsymbol{J}}$ is the total angular momentum operator.

The magnetic moment of $Tb(Cu_{0.7}Ni_{0.3})_2$ has been calculated by means of the statistical formula:

$$\boldsymbol{M} = \sum_m \langle m|\mu_B g\hat{\boldsymbol{J}}|m\rangle \exp(-E_m/k_BT)\cdot\left(\sum_m \exp(-E_m/k_BT)\right)^{-1}, \qquad (3)$$

[1)] Contribution presented at the 8th Conference on Magnetism, KOŠICE, 29. 8. 1988—2. 9. 1988

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where E_m and $|m\rangle$ are the eigenvalues and eigenvectors of the Hamiltonian (1), respectively, determined by a selfconsistent process. E_m and $|m\rangle$ are in general strongly dependent on the magnitude and the direction of $(\boldsymbol{H}_m + \boldsymbol{H})$. First we have calculated the magnetization process in the $\boldsymbol{b}$ direction at 4.2 K ($\lambda = 2.93\ T/\mu_B$) with two different sets of CEF parameters described in table 1 as experimental [2] ones and calculated by us. We have found in this case only a slow linear increase of magnetization with saturation at a high magnetic field (≈ 30 T) without any jump. Further analysis has indicated the leading role of the parameters V_4^0 and V_4^4 in the character of the crossover effect.

For this purpose we have taken the constants V_2^0, V_2^2, V_4^0 and V_4^4 as adjustable variables and have calculated the magnetization process. The other CEF constants in the Hamiltonian (2) have been neglected. The reason for this neglection is the uncertainty of such a fitting procedure of one measured property with nine CEF parameters.

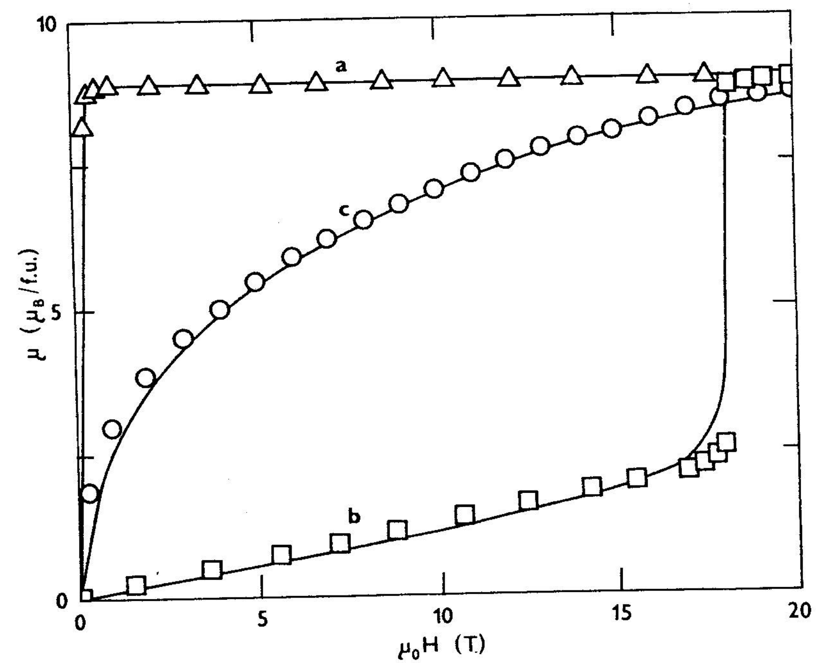

Fig. 1. Magnetization isotherms of the $Tb(Cu_{0.7}Ni_{0.3})_2$ single crystal along the principal crystallographic axes at $T = 4.2$ K. The marks represent the experimental data [4], the full lines are fits according to our model.

The obtained agreement between measured [4] and calculated magnetization curves in all three principal crystalline directions $\boldsymbol{a}$, $\boldsymbol{b}$ and $\boldsymbol{c}$ at 4.2 K can be seen in Fig. 1. The corresponding sets of CEF parameters are summarized in table 1. The splitting of the energy levels for Tb as a function of the external field along the $\boldsymbol{b}$ axis (Fig. 2) shows a discontinuous change in $H = 18$ T if we take the molecular interaction into account.

Table 1

CEF parameters for $Tb(Cu_{0.7}Ni_{0.3})_2$. The experimental values of the parameters V_2^0, V_2^2 have been obtained in the first approximation from the values of the paramagnetic Curie temperatures [2]. Our parameters are calculated in the point charge model in the sphere $r = 1.4$ nm with the shielding factor $\sigma = 0.75$ for V_2^0, V_2^2 and the fitted ones correspond to the experimental data from [4]

	V_2^0	V_2^2	V_4^0 (10^{-4})	V_4^2 (10^{-3})	V_4^4 (10^{-2})	V_6^0 (10^{-6})	V_6^2 (10^{-6})	V_6^4 (10^{-6})	V_6^6 (10^{-6})
					$(\times 10^{-23}$ J/ion)				
exp. [2]	0.98	1.59	0	0	0	0	0	0	0
calc.	1.59	1.24	4.3	−3.2	1.1	1.9	5.8	6.9	7.6
fitted	0.8	0.7	−80	0	0.9	0	0	0	0

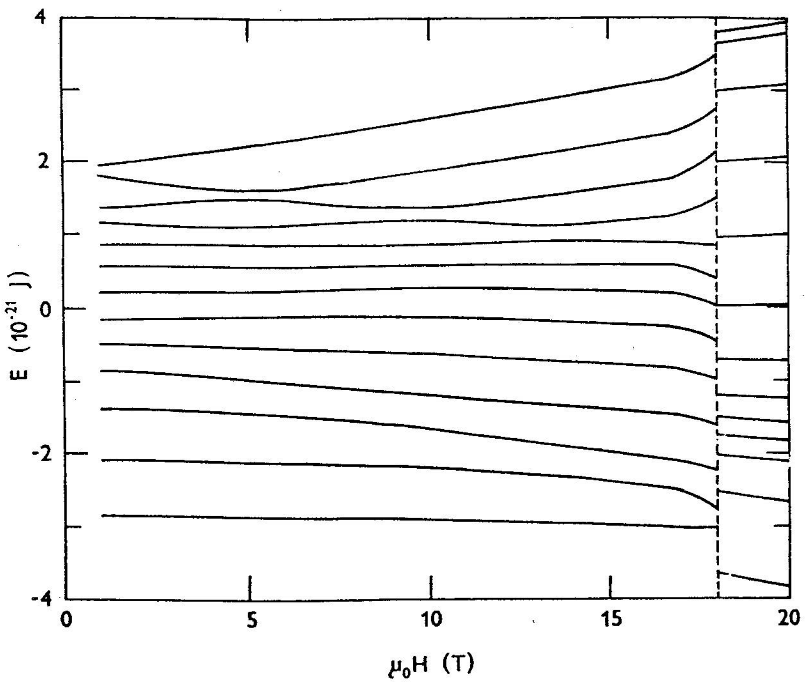

Fig. 2. The change in the CEF splitting energy levels for the $Tb(Cu_{0.7}Ni_{0.3})_2$ compound as a function of the external magnetic field along the ***b*** axis using $\lambda = 2.93\ T/\mu_B$ obtained from the paramagnetic Curie temperature $\Theta_p = 62$ K according to the equation $\lambda = 3k_B\Theta_p/g^2\mu_B^2J(J+1)$ [5].

We can conclude that the analysis of high field magnetization measurements on $Tb(Cu_{0.7}Ni_{0.3})_2$ provides the evidence that the crystal electric field Hamiltonian cannot be described sufficiently within the second order approximation $\hat{H}_{CF} = V_2^0\hat{O}_2^0(\hat{J}) + V_2^2\hat{O}_2^2(\hat{J})$ only. We must take into account no less than V_2^0, V_2^2, V_4^0 and V_4^4 parameters to obtain a satisfactory agreement with high field magnetization measurements. A further improvement of this simple CEF model is now in progress and will be published elsewhere.

REFERENCES

[1] Poldy, C. A., Gratz, E.: J. Magn. Mag. Mat. *8* (1978), 223.
[2] Hashimoto, Y.: J. Sci. Hiroshima Univ., *A43* (1979), 157.
[3] Šima, V., Smetana, Z.: Solid State Comm. *49* (1985), 981.
[4] Yumaguzhin, R. Yu., Levitin, R. Z., Popov, Yu. F.: Fizika tverdogo tela *27* (1985), 579.
[5] Shohata, N.: J. Phys. Soc. Jap., *42* (1977), 1873.

Received September 16th, 1988
Accepted for publication December 16th, 1988