Magnetic Field Induced Ising Axis Conversion in $Tb_{0.5}Dy_{0.5}Cu_2$ Single Crystals

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We study the Ising axis conversion in a mixed $Tb_{0.5}Dy_{0.5}Cu_2$ single crystal. Interest is focused on how changes in the exchange interactions due to rare earth substitutions influence the existence of magnetic phases and the critical field values for the Ising axis conversion. From magnetisation measurements we determined the (H - T) phase diagram for magnetic fields parallel to the easy a-axis and the temperature dependence of the critical field for the Ising axis conversion. Both properties for the mixed crystal follow a simple composition scaling behavior. But in contrast to previous studies on the pure compounds $TbCu_2$ and $DyCu_2$ the changes of magnetic and structural properties at the conversion cannot be recovered completely by thermal treatment. Only a small part (10 % of the sample volume) goes back to the virgin state after warming the sample to 500 K. This behavior is of great interest for further neutron or X-ray diffraction studies of the Ising axis conversion allowing to study the converted phase under routinely used experimental conditions.

I Introduction

The magnetic properties of the RCu_2 intermetallics, which crystallize (with the exception of $LaCu_2$) in the orthorhombic $CeCu_2$ structure (space group *Imma*), were intensively investigated for most members of the series. They show complex antiferromagnetic behavior, that can be modelled by anisotropy effects due to crystal field and bilinear exchange interactions [1]. Besides the complex magnetic order some of the RCu_2 compounds (R = Ce, Pr, Tb, Dy) show a change of the easy (Ising) axis in high magnetic fields [2]. The crystallographic a-axis is the easy magnetic axis of these substances in low magnetic fields. In an external field parallel to the c-axis the moments are turned steadily into the c-direction. At a critical field, however, a remarkable increase of the magnetisation occurs (compare discussion of Fig. 3) resulting in an a-axis like magnetic behavior. After ramping down the field the substance shows magnetic properties that are characteristic for fields along a in the virgin crystal. Such an irreversible magnetic transition is called "Ising axis conversion" and is thought to be caused by strong magneto-elastic interactions in the ac-plane of the compound. In a $DyCu_2$ single crystal it was demonstrated that the conversion is accompanied by a structural change from orthorhombic into hexagonal symmetry involving giant magnetostriction [3].

$Tb_{0.5}Dy_{0.5}Cu_2$ belongs to the mixed compounds of the RCu_2 series. Up to now, results were reported only for magnetically diluted mixed compounds, e.g., $Tb_xY_{1-x}Cu_2$, $Dy_xY_{1-x}Cu_2$ [4, 5]. In the present work, we report on the magnetic properties of a mixed crystal with different R^{3+} ions investigated by magnetisation and neutron diffraction.

II Experimental Details and Results

Polycristalline samples were prepared by arc-melting stoichiometric portions of the pure elements under argon atmosphere. The phase purity was checked by X-ray diffraction and the lattice parameters at room temperature have been determined (a = 4.308 $\AA$, $b = 6.816$ $\AA$, $c = 7.301$ $\AA$, $V = 214.0$ $\AA^3$). The Czochralski method was used for the $Tb_{0.5}Dy_{0.5}Cu_2$ single crystal preparation at the Forschungszentum Jülich.

Neutron powder diffraction measurements were performed on powder samples at the E6 diffractometer at the Hahn-Meitner-Institut Berlin (Germany) in zero field in order to study the crystalline and the magnetic order of $Tb_{0.5}Dy_{0.5}Cu_2$ and to compare them to the properties of the pure compounds. An Oxford MagLab2000 system, equipped with a superconducting

7 T - magnet, was used for the magnetisation measurements using an extraction magnetometer and also for the susceptibility measurements with the ac-method.

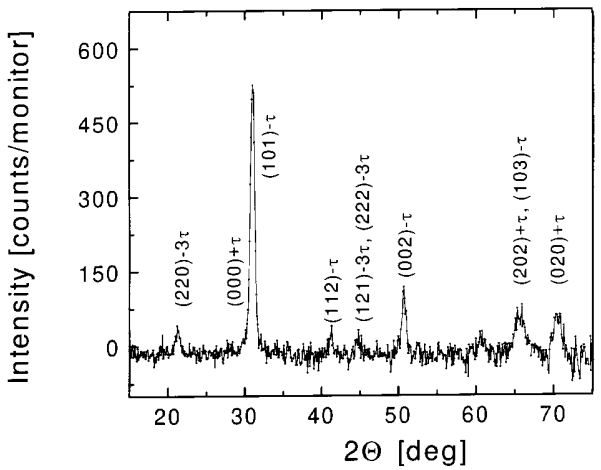

Figure 1. Neutron diffraction difference pattern of a $Tb_{0.5}Dy_{0.5}Cu_2$ powder sample at T = 1.6 K and T = 70 K (E6, $\lambda_0 = 2.42$ Å, Hahn-Meitner-Institut Berlin). The magnetic reflections with τ and 3τ are marked.

Studying the crystalline structure of $Tb_{0.5}Dy_{0.5}Cu_2$ with neutrons, no superstructure reflections have been observed. Peak-form analysis has shown Gaussian line shapes which indicates the absence of short range order. Therefore, a random distribution of the Tb and Dy atoms on the rare earth sites can be assumed. At low temperature $T = 1.6$ K the magnetic order can be described by the first and third harmonics of the wave vector $\tau = (2/3\,1\,0)$ that is also found in $TbCu_2$ and $DyCu_2$ (see Fig. 1). No additional features were observed. The saturation moment is estimated as 8.4 μ_B / f.u. Between 26 K and $T_N = 38.5$ K the reflections according to the third harmonics of τ are absent. From this we can state that in $Tb_{0.5}Dy_{0.5}Cu_2$ the magnetic structures in zero field and their temperature evolution is similar to the case of $DyCu_2$: there is a commensurate antiferromagnetic low temperature phase AF1 followed by an incommensurate, sinusoidally modulated phase AF2 [6].

The magnetic phase diagram for fields along the a-direction (the Ising axis of the system on initial cooling) was investigated by magnetisation and susceptibility measurements. The result is shown in Fig. 2. In zero field magnetic order occurs below the Néel temperature $T_N = 38$ K consistent with the value found by neutron scattering. A critical temperature of 40 K, approximately, can be estimated from T_N of pure $TbCu_2$ and $DyCu_2$ taking into account the measured saturation magnetisation M_a and the scaling of the exchange parameter $\mathcal{J}(\mathbf{Q})$ with the Landé factor g [1]:

$$kT_N = M_a^2 \mathcal{J}(\mathbf{Q} = (2/3\,1\,0)) \qquad \text{with}$$

$$\mathcal{J}(\mathbf{Q} = (2/3\,1\,0)) \propto (g-1)^2 \qquad (1)$$

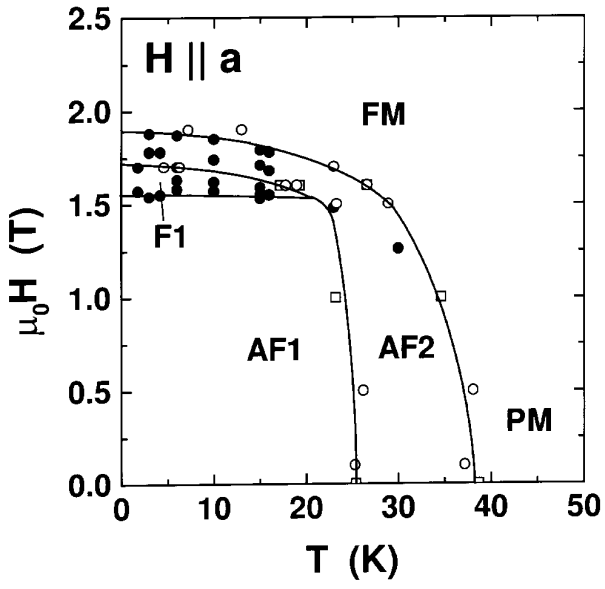

Figure 2. Magnetic phase diagram of $Tb_{0.5}Dy_{0.5}Cu_2$ (single crystal) for magnetic field in a-direction as deduced from field dependent (closed circles) and temperature dependent (open circles) magnetisation and susceptibility (open squares) measurements.

Below T_N the phase AF2 is stable down to $T_1 = 26$ K. The behavior in magnetic fields was studied both by temperature and field dependent magnetisation and susceptibility cycles in order to cross the transition lines almost perpendicularly. The phase transition points were taken from the maxima observed in these curves. Analysing the results the phase AF1 exists up to 1.55 T for $T < 25$ K. The field dependence of the transition temperature T_1 from AF1 to AF2 is rather weak. The existence of AF2 is extended to higher fields at low temperatures. But, in contrast to the pure compounds a multistep magnetisation process has been observed between 1.55 T and 1.7 T at temperatures below 20 K. Within this region, marked as F1 in the phase diagram, hysteretic effects have been observed when ramping the field up and down. From $NdCu_2$ it is known [7] that the stability of magnetic structures in the field range near the transition into the induced ferromagnetic state is extremely sensitive to small variations in the exchange constants. Similar to that behavior the long range interaction in the mixed crystal can be influenced by small deviations from the random distribution of the rare earth ions on a microscopic scale. This leads to the behavior observed in F1. At least, above 1.8 T all moments are oriented parallel to the external field and the sample is in the induced ferromagnetic state FM. The saturation moment in fields along the a-direction is about 8 μ_B / f.u. in good agreement with the value found by neutron diffraction.

We now discuss the Ising axis conversion of $Tb_{0.5}Dy_{0.5}Cu_2$ in fields along the c-direction. Magnetisation measurements were performed at different temperatures to study the effect. Two curves for $T = 30$ K

are presented in Fig. 3. The upper magnetisation curve shows a significant magnetisation jump at 3.6 T for the virgin sample. Such a jump is characteristic for the Ising-axis conversion. The absolute magnetisation after the transition correponds to the saturation moment of 8 μ_B / f.u. initially measured in fields along a-direction. All subsequent magnetisation measurements at increasing as well as decreasing fields parallel to c show the a-axis like behavior. Therefore, the existence of the Ising-axis conversion is confirmed also for the mixed crystal $Tb_{0.5}Dy_{0.5}Cu_2$. The transition field of 3.6 T near the Néel temperature T_N for crystals which were never converted before is between the minimum conversion fields of 3.2 T and 5.6 T for $DyCu_2$ and $TbCu_2$, respectively.

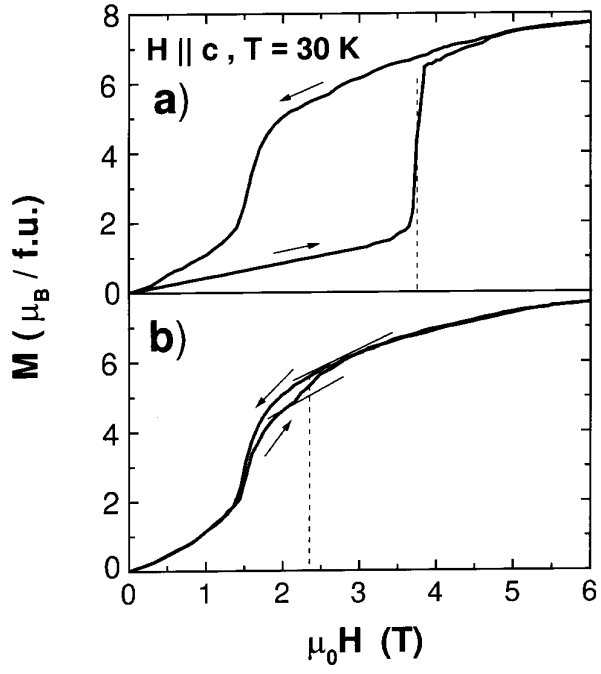

Figure 3. Magnetisation of $Tb_{0.5}Dy_{0.5}Cu_2$ (single crystal) for magnetic field in c-direction at T = 30 K indicating the Ising axis conversion (marked by dotted line): a) virgin sample A (cycle A1), b) sample B converted and afterwards annealed at 500 K (cycle B8).

In order to determine the temperature dependence of the conversion field more field cycles of the magnetisation were performed on the same crystal. To study the temperature dependence of the conversion process it is necessary to return the sample into the original state. From the pure substances it is known that a recovery of the original Ising axis (a-direction) is obtained for almost the complete sample either by thermal treatment (warming up the sample to 300 K) or by applying a magnetic field along the a-axis. But our experiments on the mixed crystals yield an important difference. From the magnetisation curves the fraction of the crystal after annealing at room temperature that still shows a-axis like behavior (called a-like part) can be estimated. This part exhibits metamagnetic transitions at fields characteristic of the a-axis (comp. phase diagram Fig. 2). Only the recovered part undergoes a new conversion. Such a two step like magnetisation curve is shown in Fig. 3 (lower part). In all measurements, the amount of the non-recovered part is about 90 %. This value does not increase significantly after more conversion cycles. In order to recover a larger part of the crystal the annealing temperature and the annealing time were increased (to 4 h at 500 K), but the effect of this treatment on the axis recovery was small. Therefore, the converted state seems to be better "locked" in the mixed crystal compared with the pure compounds. This might be caused structurally: The crystal symmetry changes from orthorhombic to hexagonal at the Ising axis conversion [3] because of the strong magneto-elastic interaction in the ac-plane. Our results suggest that the hexagonal AlB_2 structure is better stabilized if different R^{3+} ions are present.

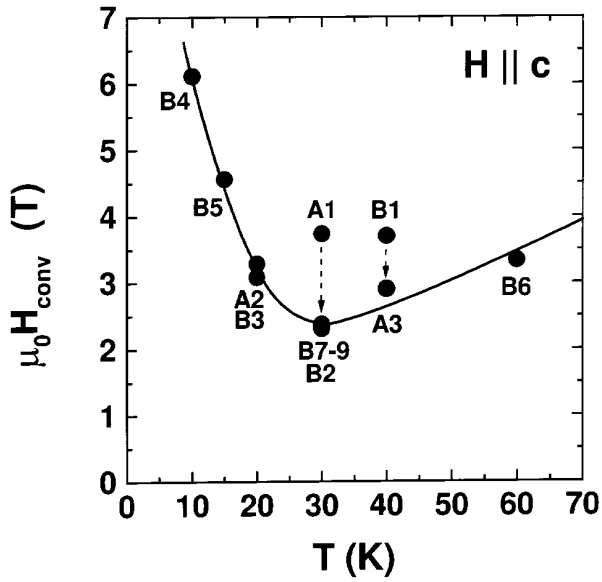

Figure 4. Ising axis conversion fields for $Tb_{0.5}Dy_{0.5}Cu_2$ (two single crystals, A and B). The numbers indicate the number of the conversion cycle. Between the conversion cycles the samples were annealed at 300 K (A1-A3, B1-B6 and B9), 400 K (B7) and 500 K (B8). The reduction of the conversion field after the first conversion cycle (shown by arrows) is explained by an incomplete relaxation (see text).

The temperature dependence of the conversion field for the two investigated samples (A and B) is presented in Fig. 4 for several conversion cycles. The shape of the curve is similar to that of the pure compounds [6, 8]: The conversion field decreases with temperature in the low temperature region, because the long range magnetic order stabilises the non-converted state, and reaches a minimum of 2.5 T just below the ordering temperature. Then it increases almost linearly due to the quadrupolar interaction [2]. Moreover, it was found that the insufficient relaxation of the crystal leads to a remarkable decrease of the conversion field. The

reduction of the conversion field after the first cycle can be seen by comparing different conversion cycles on the same sample. Both the fact that almost the whole sample remains converted after handling it at room temperature, and the reduction of the conversion fields open new opportunities to study the converted state. Routinely used experimental methods (e.g. X-ray scattering for investigation of crystalline structure, neutron scattering to analyse magnetic structure and excitations etc.) now are possible starting with a once converted crystal.

In summary, we investigated the magnetic behavior of a $Tb_{0.5}Dy_{0.5}Cu_2$ mixed crystal. The rare earth ions Tb^{3+} and Dy^{3+} seem to be almost randomly distributed. The conversion of the magnetic Ising axis in external fields along the c-axis also exists in this substance. But, in contrast to the behavior of $TbCu_2$ and $DyCu_2$ the system cannot be relaxed by thermal treatment at temperatures up to 500 K. This indicates that the energy that is necessary for a recovery of the original state is larger than in the pure compounds.

Acknowledgements

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References

[1] M.Rotter, M.Loewenhaupt, S.Kramp, T.Reif, N.M. Pyka, W.Schmidt and R.Kamp, *Europ. Phys. J.* **B 14** (2000) 29

[2] K. Sugiyama, M. Nakashima, Y. Yoshida, R. Settai, T. Takeuchi, K. Kindo and Y. Onuki, *Physica* **B 259-261** (1999) 896

[3] P. Svoboda, M. Doerr, M. Loewenhaupt, M. Rotter, T. Reif, F. Bourdarot and P. Burlet, *Europhys. Lett.* **48** (1999) 410

[4] N. H. Luong, T. D. Hien and N. H. Duc, *Physica B* **109 & 110** (1982) 2135

[5] T. D. Hien, N. H. Luong and N. H. Duc, *J. Magn. Magn. Mat.* **31-34** (1983) 245

[6] M. Loewenhaupt, M. Doerr, L. Jahn, T. Reif, C. Sierks, M. Rotter and H. Mueller, *Physica* **B 246-247** (1998) 472

[7] M. Rotter, M. Loewenhaupt and S. Kramp, *Physica* **B 276-278** (2000) 598

[8] M. Doerr, M.Rotter, M. Loewenhaupt, T. Reif and P. Svoboda, *Physica* **B** 276-278(1999) 1331