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To cite this version:

V. Tsurkan, S. Radautsan, M. Baran, R. Szymczak, H. Szymczak. Magnetization Anomalies in some Ternary Spinel Single Crystals. Journal de Physique IV Proceedings, 1997, 07 (C1), pp.C1-271-C1-272. 10.1051/jp4:19971106 . jpa-00254727

HAL Id: jpa-00254727

https://hal.science/jpa-00254727v1

Submitted on 4 Feb 2008

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J. PHYS. IV FRANCE 7 (1997)
Colloque C1, Supplément au Journal de Physique III de mars 1997

Magnetization Anomalies in some Ternary Spinel Single Crystals

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Abstract. DC magnetization studies of the $Cu_{1+v}Cr_2Se_{4-x}Br_x$ and $Hg_{1-x}Cu_xCr_2Se_4$ magnetic spinel single crystals for different substitutions that induce bond and site disorder, revealed the reentrant spin-glass-like character of magnetization and the noticeable influence on the phase transition.

I. INTRODUCTION

Magnetic chromium chalcogenides ACr_2X_4 (A= Cd, Hg, Cu, Zn; X= S, Se) exhibit a variety of magnetic structures depending on the strengths of competing ferro- and antiferromagnetic exchange interactions. Substitution and doping can induce fluctuations of these interactions and influence on the phase transition. In some cases, this disorder can cause the spin-glass or cluster-like behavior of magnetization [1 - 5].
Here we present the results of detailed investigation of the influence of disorder on the magnetic properties of $Cu_{1+v}Cr_2Se_{4-x}Br_x$ and $Hg_{1-x}Cu_xCr_2Se_4$ single crystals previously observed in [6-8].

2. EXPERIMENTAL

$Cu_{1+v}Cr_2Se_{4-x}Br_x$ single crystals were grown by flux method from the CuBr and $CuBr_2$ melts. $Hg_{1-x}Cu_xCr_2Se_4$ single crystals were obtained by chemical vapour transport reactions. Sample composition has been checked by EPMA and EDAX analyses with an ~1- 2 % errors for cations and 2-5 % for anions. Magnetic measurements have been performed using vibrating sample magnetometer (PAR- 155) and SQUID magnetometer (Quantum Design MPMS- 5) in 4.2- 300 K temperature range and magnetic field intensity up to 1.6 T and 5T, respectively.

3. EXPERIMENTAL RESULTS

All the studied crystals exhibit spin-glass- like behaviour of magnetization at low temperatures: difference between field cooled (FC) and zero field cooled (ZFC) magnetizations, irreversible remanent magnetization and other peculiarities dependent on the thermomagnetic history of the samples.
A sharp decrease of ZFC magnetization with the further decrease of the temperature has been observed in Cu-rich $Cu_{1+v}Cr_2Se_{4-x}Br_x$ samples after the increase of magnetization due to the transition from paramagnetic (PM) to ferromagnetic (FM) state [6]. The increase of the magnetic field intensity was found to reduce the value of this lowering and to shift the temperature of FM to spin-glass-like transition T_f to the lower temperatures (Fig.1).
Thermoremanent magnetization (TRM) has been found to consist of two parts: the first one comes from infinite ferromagnetic matrix and the second one originates from the frustrated subsystem. The value of the latter has the nonmonotonous magnetic field dependence and a maximum for about 200 Oe (Fig.2). The value of TRM were observed to depend on the cooling history and the irreversible part decreases with the increase of the reversible TRM.
The shifted FC hysteresis loops have been observed for these samples.
The saturation magnetization M_s of these crystals shows the significant deviation from the $T^{3/2}$ law at low temperatures.
In $Hg_{1-x}Cu_xCr_2Se_4$ single crystals the spin-glass- like behaviour of magnetization at low temperatures has been also observed [8]. The values of the difference between ZFC and FC magnetizations as well as TRM were found to increase with the substitution concentration. The similar effects have been also observed for crystals doped with Au or Ag.
It should be pointed out that undoped $HgCr_2Se_4$ crystals as well as crystals doped with In or Ga for the same concentration range ($x < 0.05$) do not show any similar peculiarities of magnetization at low temperatures.
In both systems the magnetization studies near T_C have been performed because of the substantial influence of the Cu ions on the temperature of PM- FM phase transition.
In $Cu_{1+v}Cr_2Se_{4-x}Br_x$ crystals the linearization of the modified Arrott plots in the asymptotic region has been obtained with the values of the critical exponents $\beta = 0.5$ and $\gamma = 1.59$. The third critical parameter δ determined from the dependence

Article published online by EDP Sciences and available at http://dx.doi.org/10.1051/jp4:19971106

ln M= f(lnH_{eff}) has the value 4.8 ± 0.2.
For $Hg_{1-x}Cu_xCr_2Se_4$, near the Curie temperature, the modified Arrott plots with the values of the critical exponents β and γ typical for 3d Heisenberg model have been found to be non-linear in high magnetic fields for undoped crystals. This peculiarity becomes less pronounced with the increase of the Cu concentration in crystals.

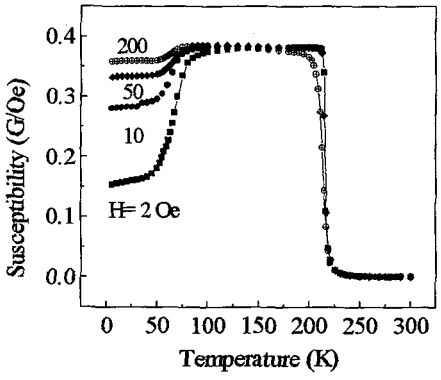

Fig.1. ZFC susceptibility vs. temperature for $Cu_{1+v}Cr_2Se_{4-x}Br_x$ crystal (v = 0.24, x= 0.6)

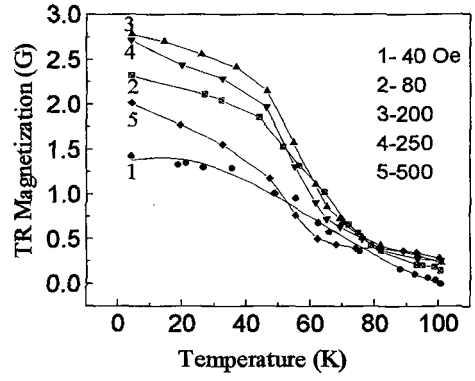

Fig.2. Thermoremanent magnetization of frustrated subsystem vs. temperature for FC in different magnetic fields (sample of fig.1).

4. DISCUSSION

The obtained data point to the substantial influence of the disordering induced by the cation substitution on the magnetic properties of systems studied.
The similar results observed for Cu-deficient $CuCr_2Se_{4-x}Br_x$ samples were related to the increase of the bond disorder with the anion substitution due to the increase of the fluctuation of the positive Cr-Se (Br)-Se (Br)- Cr superexchange interactions [6]. In $Cu_{1+v}Cr_2Se_{4-x}Br_x$ system, the excess v Cu ions, if they occupy interstitial tetrahedral sites like those in $Cu_{1+v}Cr_2Te_4$ crystals [9], can increase the number of negative Cr-Se (Br)- Cu- Se (Br)- Cr interactions in crystals and thereby induce the fluctuations of superexchange interactions or site disorder. Taking into account the low experimental value of the total magnetization, it cannot be excluded that some part of Cu ions may occupy the octahedral sites in $3d^9$ configuration with the magnetic moment antiparallel to that of chromium ions. This incorporation can also result in the dilution of the Cr subsystem and contribute to the frustration phenomena in a way similar to that in reentrant spin-glass $CdCr_{2-2x}In_{2x}S_4$ samples [2].
The fact that in $Hg_{1-x}Me_xCr_2Se_4$ system we have observed the spin-glass like behaviour of magnetization only in crystals with acceptor substitution (Me= Cu^+, Ag^+, Au^+) could point to the influence of the magnetic ions with the strong spin-orbital coupling (Cr^{4+}) on the frustrated states.
Regarding to the influence of the substitution on the phase transition it should be mentioned that the value of the critical parameter γ obtained for $Cu_{1+v}Cr_2Se_{4-x}Br_x$ crystals indicates relatively strong disorder, whereas the other critical exponent β has the mean field value. A slight disagreement between the values of γ and β and the value of δ determined independently is probably connected with the above mentioned nonlinearity of the modified Arrott plots.
It should be also mentioned that the behaviour of the critical exponents for $Hg_{1-x}Cu_xCr_2Se_4$ crystals is difficult to explain in terms of the existing notions of the influence of the disorder on the critical phenomena of the ferromagnets [10].

References

[1] Dormann J.L., Nogues M., Phase Trans. **33** (1991) 159.
[2] Alba M., Hammann J., Nogues M., J. Phys. C: Solid State Phys. **15** (1982) 5441.
[3] Pouget S., Alba M., Nogues M., J. Appl. Phys. **75** (1994) 5826.
[4] Koroleva L.I., Virovets T.V., phys. stat. sol. (b) **157** (1990) 431.
[5] Plumier R., Sougi M., Solid State Commun. **69** (1989) 341.
[6] Baran M., Szymczak R., Szymczak H., Tsurkan V., J. Magn. Magn. Mater. **140- 144** (1995) 2043.
[7] Radautsan S., Tsurkan V., Baran M., Szymczak R., Szymczak H., J. Crystall Res. and Technology **31** (1996) 579.
[8] Tsurkan V., Tezlevan V., Muscutariu I., Baran M., Szymczak R., Kiosse Gh., Petrenko P., J. Crystall Res. and Technology **31** (1996) 639.
[9] Lotgering F.K., van der Steen G.H.A.M., Solid State Commun. **9** (1971) 1741.
[10] Kaul S.N., J. Magn. Magn. Mater. **53** (1985) 5.