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MAGNETIC STRUCTURE OF Cu$_x$Zn$_{1-x}$Cr$_2$Se$_4$

S. Niziol (1), A. Bombik (2), D. Fruchart (3), J. Kusz (4) and J. Warczewski

(1) Laboratory of Neutron Physics, JINR, Head Post Office, Box 79, 1010000 Moscow, U.S.S.R.
(2) JPNT AGH, Cracow, Poland
(3) C.N.R.S., Grenoble, France
(4) Silesian University, Katowice, Poland

Abstract. – Magnetic and neutron diffraction measurements on Cu$_x$Zn$_{1-x}$Cr$_2$Se$_4$ yield a simple spiral structure for $x = 0.02$ and a conical one for $x = 0.11$ below 21 K. Cu$_{0.11}$Zn$_{0.89}$Cr$_2$Se$_4$ proved to be of the magnetic cluster glass type in contradiction with theoretical predictions of Krok et al. for 0.05 ≤ $x$ ≤ 0.2 [5].

Introduction

The spinels Cu$_x$Zn$_{1-x}$Cr$_2$Se$_4$ exhibit rather complicated magnetic properties. On one end of the series there is ferromagnetic CuCr$_2$Se$_4$ [2], on the other helimagnetic ZnCr$_2$Se$_4$ (below $T_N = 21.2$ K; neutron diffraction data [1]). In a number of papers [1-5] it has been shown that it is the presence of the Cu atoms that controls onset of a particular magnetic ordering within the system. Cu is responsible for the formation of the Cr$^{4+}$ ions which enter into the magnetic double exchange interaction with Cr$^{3+}$. Thanks to the theoretical calculations of [5], there exists a description of how the Cu concentration influences such an interaction in Cu$_x$Zn$_{1-x}$Cr$_2$Se$_4$ and how it controls the transition from helimagnetism for $x = 0.0$ through a conical structure to ferromagnetism for $x ≥ 0.8$.

For 0.1 ≤ $x$ ≤ 0.2 there is a remarkable jump of the critical temperature of the magnetic ordering: the Néel temperature $T_N = 21$ K for $x = 0.1$ evolves into the Curie temperature $T_C = 377$ K for $x = 0.2$ [3]. On the other hand, theory [5] predicts a continuous change of the spiral and conical angles ($\varphi(x)$ and $\psi(x)$) within this region of the Cu concentration. Up to now, this prediction could not be verified due to the lack of sufficient experimental data.

It is the aim of this paper to provide such data through the measurements of neutron diffraction spectra and magnetic susceptibility of powdered samples: the latter in a constant low magnetic field.

Experimental results

MAGNETIC PROPERTIES. – Magnetic susceptibility of Cu$_{0.11}$Zn$_{0.89}$Cr$_2$Se$_4$ has been measured within the temperature range of 7 K – 300 K, using a magnetic balance of high sensitivity. Taking into account the jump of the magnetic ordering temperature [5] for 0.1 ≤ $x$ ≤ 0.2 as well as the change of the Néel temperature in an applied magnetic field for 0.0 ≤ $x$ ≤ 0.1 [2], it was decided to carry out the measurements for the field-cooled samples. The temperature dependence of reciprocal susceptibility $\chi^{-1}(T)$ in a low temperature region for various values of the applied field is shown in figure 1. Curve 1 refers to $\chi^{-1}(T)$ for the sample cooled from 300 K to 7 K without any magnetic field. A rather wide minimum due to the change of the magnetic ordering can be seen. Curves 2, 3 and 4 present $\chi^{-1}(T)$ for the samples cooled in the applied constant fields $H_C = 0.034$ T, $H_C = 0.058$ T and $H_C = 0.13$ T, respectively. The curves 2 and 3 have been taken in the same field that was used to provide the needed cooling conditions, i.e. the field was neither switched off nor changed before the completion of the measurement. The measurements for curves 1 and 4 have been performed in the constant field $H = 0.022$ T. The shape $\chi^{-1}(T)$ apparently changes for the field-cooled samples, and the temperature of magnetic phase transition becomes more precisely defined (∼ 21 K). However, with increasing $H_C$, the minimum of $\chi^{-1}(T)$ becomes more shallow and probably vanishes for $H_C$ of

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about 0.13 T. Above 40 K, the dependence of $\chi^{-1}(T)$ upon $H_c$ is no longer seen. Besides, with the increasing temperature, the $\chi^{-1}(T)$ curves become steeper so that the existence of another phase transition is hardly possible.

MAGNETIC STRUCTURE. — Neutron scattering experiments were carried out for both samples ($x = 0.02$ and $x = 0.11$) at the temperatures of 4.8 K and 80 K for $x = 0.02$, and at 4.8 K, 9.3 K, 13.9 K, 23 K, in the case of $x = 0.11$. For both compounds the neutron diffraction patterns obtained at low temperatures ($T < 23$ K) contain peaks typical for a spinel structure (Fd3m) plus a number of magnetic satellites. The intensity of the satellites decreases with increasing temperature and becomes zero at about 21 K.

Both samples ($x = 0.02$ and $x = 0.11$) appear to be normal spinels with the oxygen parameter $u = 0.385$ and the following lattice constants at 4.8 K: $a = 10.445$ Å ($x = 0.02$) and $a = 10.432$ Å ($x = 0.11$). From the positions of satellites, the propagation vectors $Q$ of modulated structures could be determined. They have been found parallel to one of the (100) axes.

At 4.8 K Cu$_{0.02}$Zn$_{0.98}$Cr$_2$Se$_4$ has a simple spiral magnetic structure of the ZnCr$_2$Se$_4$ type [1] with $Q = 0.42$, $\varphi = 38^\circ$, and $\mu_{Q+1} = 1.75\ \mu_B$.

The results for Cu$_{0.11}$Zn$_{0.89}$Cr$_2$Se$_4$ are a bit more complicated. Certain reflections are somewhat broadened in this case (Fig. 2 and 3). The broadened structural peaks can be decomposed into two parts, one of them being of a diffusion origin with temperature-dependent intensity (that did not vanish above 21 K). (see Fig. 3). One may assume that Cu$_{0.11}$Zn$_{0.89}$Cr$_2$Se$_4$ is built of small regions with a conical magnetic ordering. Under this assumption we have estimated the structural parameters (see Tab. I). The diffusional components, von-vanishing above the 21 K phase transition, may prove the existence of ferromagnetic order-

![Fig. 2. - Diffraction pattern of Cu$_{0.11}$Zn$_{0.89}$Cr$_2$Se$_4$ at 4.8 K in proximity to the peak (111).](image1)

![Fig. 3. - Diffraction pattern of Cu$_{0.11}$Zn$_{0.89}$Cr$_2$Se$_4$ at 23 K in proximity to the peak (111).](image2)

![Table I. - At $T = 0$ K; $\varphi = 35^\circ$; $\nu = 60^\circ$ in reference [5].](image3)

<table>
<thead>
<tr>
<th>$T$ [K]</th>
<th>$Q_x$</th>
<th>$\varphi^0$</th>
<th>$\nu^0$</th>
<th>$\mu_{Q} [\mu_B]$</th>
<th>$\mu_{Q+1} [\mu_B]$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.8</td>
<td>0.448</td>
<td>40.3</td>
<td>29</td>
<td>1.75</td>
<td>0.95</td>
</tr>
<tr>
<td>9.3</td>
<td>0.447</td>
<td>40.1</td>
<td>29</td>
<td>1.75</td>
<td>0.95</td>
</tr>
<tr>
<td>13.9</td>
<td>0.443</td>
<td>39.9</td>
<td>22</td>
<td>1.80</td>
<td>0.75</td>
</tr>
<tr>
<td>23</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1.85</td>
</tr>
</tbody>
</table>

It can be noticed that Cu$_{0.11}$Zn$_{0.89}$Cr$_2$Se$_4$ does not become paramagnetic at 21 K. At a temperature 295 K the coherent magnetic scattering was not observed for this sample.

Both the neutron diffraction results and the $\chi^{-1}(T)$ for the field-cooled samples are similar to those obtained for materials known as magnetic cluster glasses. Hence, it can be inferred that this is also the case for Cu$_{0.11}$Zn$_{0.89}$Cr$_2$Se$_4$.

We would like to conclude with the statement that the model worked out in [5] cannot be considered satisfactory for concentration $0.05 \leq x \leq 0.2$.