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MANGANESE IMPURITIES AND SPIN GLASS LIKE PROPERTIES IN CuZr GLASSY METALS

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Résumé.- Nous présentons une étude des propriétés magnétiques du système amorphe (Cu<sub>60</sub>Zr<sub>40</sub>)<sub>1-c</sub>Mn<sub>c</sub> (c ≤ 10 at %) dans le régime dilué et dans le régime où les impuretés magnétiques interagissent entre elles. Les caractéristiques des impuretés de manganèse dans le régime dilué sont comparées à celles obtenues dans un alliage cristallin équivalent ; dans le régime plus concentré la portée des interactions entre impuretés est discutée.

Abstract.- We present a study on the magnetic properties of the amorphous (Cu<sub>60</sub>Zr<sub>40</sub>)<sub>1-c</sub>Mn<sub>c</sub> system (c ≤ 10 at %) in the dilute regime and in the interacting regime of magnetic impurities. The characteristics of manganese impurities obtained in the dilute regime are compared to those obtained in the crystalline counterpart ; in the more concentrated regime the range of the interaction between magnetic impurities is discussed.

In this paper we study the magnetic properties of amorphous (Cu<sub>60</sub>Zr<sub>40</sub>)<sub>1-c</sub>Mn<sub>c</sub> alloys for c ≤ 10 at %; the properties of a (Cu<sub>59</sub>Zr<sub>41</sub>)<sub>99</sub>Mn<sub>1</sub> crystalline alloy are also studied. The magnetic state of manganese atoms and their long range interactions are considered. Several studies has been recently carried out on CuZr glassy metals, in particular concerning the atomic structure. However, the conclusions of the authors are not in agreement with each other /1,2/. On another hand it has been shown that a single phase exists for crystalline Cu<sub>59</sub>Zr<sub>41</sub>/3/ ; its structure is very close to that of Ni<sub>10</sub>Zr<sub>7</sub>, which presents 4 zirconium sites and 5 nickel sites /4/. Photoemission studies /5,6,7/ on CuZr and other Zr based glassy metals show in particular that the electronic structure can be understood in a band model where the Zr d band lies near the Fermi level meanwhile the Cu d band lies at the bottom of the valence band. The effect of transition impurities in a CuZr matrix has been studied /8/ ; only Mn atoms carry a localized moment which has been evaluated to 1.6μ<sub>B</sub>. Another investigation /9/ concludes to the occurence of a para-ferromagnetic transition in the Cu<sub>60</sub>-Zr<sub>40</sub>Fe<sub>5</sub> system. Let us note that manganese atoms in crystalline Cu and Zr respectively, carry their full moment ; spin glass properties are observed for appropriate Mn concentrations /10,11/.

Experimental procedure :

The amorphous alloys has been prepared by the rapid quenching technique. The X-ray analysis showed that the amorphous phase can be obtained for c up to 10 at %. The manganese concentrations have been analyzed by atomic absorption spectrometry. Specimens of Cu<sub>59</sub>Zr<sub>41</sub> and (Cu<sub>59</sub>Zr<sub>41</sub>)<sub>99</sub>Mn<sub>1</sub> were prepared as described by Lars Bsenko /3/. The magnetization measurements /12/ have been carried out by the Faraday method for fields up to 52 kOe and temperatures between 1.7 and 300 K.

Experimental results :

For all the investigated samples, the initial susceptibility χ<sub>0</sub> can be separated in a temperature independent term χ′ and a temperature dependent one which varies according to a Curie-Weiss law. The temperature independent term represents the contribution of the host and of the eventual non magnetic impurities. For glassy Cu<sub>60</sub>Zr<sub>40</sub>, χ′ is of 0.5 x 10<sup>-5</sup> emu/g ; the interpretation of such a va-
value can be found in ref. 5. With increasing manganese concentration, $\chi'$ first increases linearly then saturates progressively (Table I). This variation of $\chi'$ can be attributed to the host or more likely to the presence of non magnetic impurities. For the temperature dependent term which is attributed to manganese impurities, all the $\theta$ values are positive (Table I); as a function of manganese concentration, they first decrease ($\theta$ equals zero for $c = 3$ at %), then they increase with $c$. The determined Curie constants (between 35 and 400 $10^{-6}$ emu/g) vary linearly with concentration for $c \leq 2$ at %, then they saturate progressively and seem to decrease for 10 at %. The Curie constant for the crystalline $(Cu_{59}Zr_{41})_{99 Mn_1}$ is noticeably higher ($110 10^{-6}$ emu/g) than for the corresponding amorphous alloy. At low temperatures ($T < 20$ K) the magnetization shows a continuous approach to saturation with increasing magnetic field; this approach becomes slower for high manganese concentrations. Moreover, at a given temperature and magnetic field, the magnetization versus $c$ varies linearly for low manganese concentrations ($c \leq 2$ at %), then it saturates progressively and finally seems to decrease for $c > 7$ at % (Fig. 1). In other words, $(H,T)/c$ is an unique function of $H$ and $T$ for $c \leq 2$ at %. Deviations from this unique function are observed at higher concentrations.

In the crystalline alloy, the approach to saturation is the same as for dilute amorphous alloys (Fig. 2); only some differences can be observed at 1.7 K. However, the value of the magnetizations are noticeably higher than for the amorphous corresponding alloy.

The previous results show clearly that for the magnetic impurities at least two concentration regimes can be defined; first the dilute one where

<table>
<thead>
<tr>
<th>$c$ at %</th>
<th>$\chi'$ ($10^{-6}$ emu/g)</th>
<th>$\theta$ K</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.5</td>
<td>3</td>
</tr>
<tr>
<td>0.5</td>
<td>0.55</td>
<td>3</td>
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<tr>
<td>2.0</td>
<td>0.75</td>
<td>1</td>
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<td>3.0</td>
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<tr>
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<td>3</td>
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<td>7.0</td>
<td>1.5</td>
<td>7</td>
</tr>
<tr>
<td>10.0</td>
<td>1.5</td>
<td>7</td>
</tr>
<tr>
<td>1.0 cryst.</td>
<td>0.85</td>
<td>2</td>
</tr>
</tbody>
</table>

Fig. 1: Amorphous $(Cu_{59}Zr_{41})_{99 Mn_1}$ variation of the magnetization vs manganese concentration.

Fig. 2: Approach to the saturation of amorphous and crystalline CuZr alloys with 1 at % of manganese impurities.
Dilute concentration range \( c \lesssim 2 \) at \%:

For \( T \geq 4.2 \text{ K} \) the magnetization as a function of magnetic field and temperature can be described by the following equation:

\[
M(H,T) = M_o(H/T + \theta) + \chi'H
\]

where \( \chi' \) is the same as that deduced from the susceptibility analysis. In the low temperature limit \((T = 1.7 \text{ K})\) some deviations from the previous law are observed. This suggests that the magnetic impurities have characteristic temperatures which are of the same order of magnitude as the considered ones \( /13/ \). Let us now evaluate the magnetic moment of the impurities. The analysis of \( M_o(H/T + \theta) \) shows that it varies approximately as a Brillouin function with \( g = 2, J = 2.2 \) (i.e. \( \mu = 4.9 \beta_b/p \)). The concentration \( C_0 \) of these impurities suggests that only one Mn atom out of about \( n \) is magnetic; \( n \) equals 6.5 for the amorphous alloys and 4.5 for the crystalline one.

Interacting concentration range and spin glass like regime: \( C > 2 \) at \%:

Since for the spin glass term the definition varies with the authors, let us note that we adopt the definition proposed by Tholence and Tournier \( /14,16/ \) i.e. alloys where the magnetization and the specific heat vary according to a scaling law. In the present case, the magnetization of the impurities satisfies to such a law for \( 3 \) at \% \( \leq C \leq 7 \) at \% i.e. \( (M(H,T) - \chi'H)/c \) is an unique function of \( H/c \) and \( T/c \) for \( T/c \leq 1.4 \text{OK/at\%} \). This suggests that at low temperature, RKKY type \( (1/c^3) \) interactions occur between magnetic impurities. It is interesting to notice that the spin glass like properties occur in a concentration range where the magnetic impurities are less than 1 at \%.

Conclusions:

The magnetic properties of crystalline \((\text{Cu}_{59}\text{Zr}_{41})_{99}\text{Mn}_1\) can be discussed in terms of local environment \( /12,17/ \) of the manganese atoms. The fact that only one Mn atom out of 4.5 is magnetic suggests that at least two kinds of local environments are present in the compound. The magnetic properties of the amorphous alloys could be understood in the same way i.e. the existence of various local environments in the alloy; however some disordering effect occurs in the amorphous state since for a same Mn concentration the number of magnetic impurities is less for the amorphous alloy.

The occurrence of long range interactions between magnetic impurities gives rise to an important question about the mean free path of conduction electrons in the amorphous alloys. As a matter of fact, interactions are already detectable for concentrations of magnetic impurities of about 0.4 \%. This seems to indicate that the mean free path in an amorphous alloy can equal several interatomic distances.

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References


