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MAGNETIC PROPERTIES OF AMORPHOUS MANGANESE ALUMINOSILICATES : SPIN-GLASS LIKE BEHAVIOUR AT LOW TEMPERATURE

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Résumé.— On a mesuré la susceptibilité magnétique à 70 Hz en champ nul et en champ appliqué, l'aimantation en fonction du champ et l'aimantation rémanente d'aluminosilicates amorphes $\text{MnO} \cdot \text{Al}_2\text{O}_3 \cdot \text{SiO}_2$ de concentration atomique en Mn 18,9 % et 15,1 %, aux températures supérieures à 1,2 K. Malgré une forte interaction antiferromagnétique moyenne entre ions Mn^{2+} , les propriétés magnétiques sont très similaires à celles des alliages dits "verres de spin".

Abstract.— AC susceptibility in zero and in applied magnetic field, magnetization versus applied field and remanent magnetization of aluminosilicate glasses $\text{MnO} \cdot \text{Al}_2\text{O}_3 \cdot \text{SiO}_2$ with atomic Mn concentrations 18.9 % and 15.1 %, have been studied down to 1.2 K. In spite of the large mean value of antiferromagnetic interactions between Mn^{2+} ions, the magnetic properties are very similar to those of spin-glass alloys.

Recent measurements of Verhelst et al. [1] have shown that cobalt and manganese aluminosilicate exhibit a relatively sharp peak in the low field AC susceptibility, very similar to that observed for the spin-glass alloys as Au-Fe [2]. In order to obtain a more detailed comparison, we have extended this work on amorphous manganese aluminosilicates and performed detailed measurements which are now classical for spin-glass alloys : effect of applied fields on the AC susceptibility; high field magnetization and remanent magnetization measurements.

Two samples 1 and 2 of $\text{MnO} \cdot \text{Al}_2\text{O}_3 \cdot \text{SiO}_2$ glasses of good optical quality with respective atomic Mn concentrations 18.9 % and 15.1 % have been prepared following the method previously described [3]. Most of the experiments were performed on the sample 1.

AC SUSCEPTIBILITY : AC susceptibilities were measured using a mutual inductance bridge operating at 70 Hz. The amplitude of the AC field was kept below 5 Oe. Measurements were done at room temperature, 77 K and in the range 1.2 - 4.2 K.

From the high temperature measurements, we found respective Curie-Weiss temperatures $\theta = -100$ K and -130 K for samples 1 and 2. Measured susceptibilities versus temperature in the range 1.2 - 4.2 K are shown in figure 1. Peaks are observed at respective temperature $T_M = 3.23$ K and 2.25 K for samples

1 and 2. An additional small static field of a few hundred oersteds produces a net rounding of the peaks as for the Au-Fe alloys [2]. The decrease of the maximum susceptibility value is about 5 % for sample 1 and 2 % for sample 2 at 340 Oe.

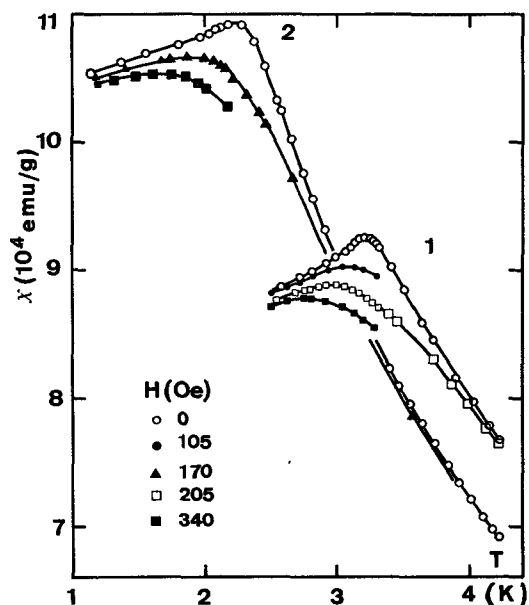


Fig. 1 : The AC susceptibility of samples 1 (18.9 Mn at %) and 2 (15.1 %) versus temperature and the effect of weak additional field.

In addition, AC susceptibility has been measured versus static field H up to 36 kOe at three fixed temperatures : 1.4 K, 4.2 K and the peak temperature T_M . The susceptibility strongly decreases when increasing field. Above 10 kOe, the $\chi(H)$ curves are identical for the three selected temperatures. Below 10 kOe, $\chi(H)$ depends on temperature and exhibits the sharpest decrease for $T = T_M$.

MAGNETIZATION VERSUS FIELD : The variation of the magnetization M with the applied field H has been deduced from Faraday rotation measurements for sample 1 (figure 2). In the present case, the rotation angle θ can be expressed as $\theta = aM + bH$. The a and b coefficients are temperature independent and have been determined from low field magnetization curves at 295 K and 4 K.

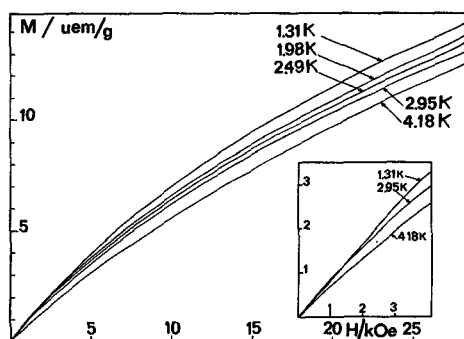


Fig. 2 : Magnetization of the sample 1 versus applied field around T_M .

The most striking features are the low field behaviour of the magnetization for $T \ll T_M$ (figure in insert) and the high field magnetization much smaller than its calculated saturation value $M_0 = 203$ emu/g. Hysteresis curves very similar to those of spin-glass alloys such as YFe_2 [4] were obtained for zero or in-field cooling through T_M .

REMANENT MAGNETIZATION : Remanent magnetization below T_c was measured by a fluxmetric method and by Faraday rotation. The time variation of the thermoremanent magnetization [5] (TRM) of sample 1 at 1.35 K is shown in figure 3. Between $t = 3$ s and 10^4 s the TRM is a linear function of $\log t$. The slope of the $\log t$ line clearly depends on applied field for small field values.

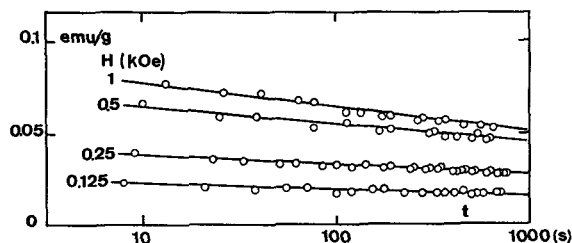


Fig. 3 : Thermoremanent magnetization of the sample 1 at 1.35 K for a series of applied field values, as a function of time.

Similar features were observed for the isothermal remanent magnetization (IRM). Figure 4 shows TRM and IRM measured at $t = 100$ s and 1.35 K, versus magnetic field. The IRM curve is below the TRM one which reaches its saturation value for relatively small fields of about 1 kOe. This saturated TRM value varies strongly with temperature and cannot be detected for $T > 2.4$ K which corresponds to about $0.75 T_M$. In the temperature range 1.2 - 2.4 K, it can be fitted by the law

$$M_{rs}(T) = 1.7 \exp(-2.5/T) \text{ emu/g.}$$

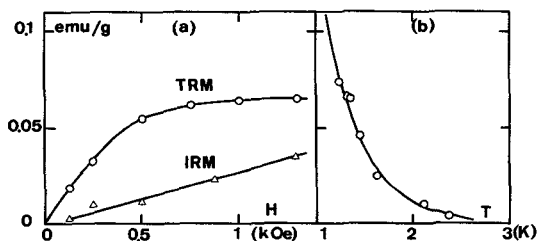


Fig. 4 : a) Field dependence of the thermoremanent and the isothermal remanent magnetization of the sample 1 measured at $t = 100$ s and $T = 1.35$ K.

b) Saturated remanent magnetization at $t = 100$ s versus temperature $M_{rs}(T)$ and the empirical law $1.7 \exp(-2.5/T)$ (solid curve).

DISCUSSION : Our θ and T_M values are about two times smaller than those Verhelst et al. This may be due to the rather different sample preparation. Since superexchange is very sensitive to the angle of Mn-O-Mn bonds, slight structural changes may strongly affect the magnetic properties. The small value of high field magnetization compared to M_0 is consistent with strong antiferromagnetic mean interactions indicated by $|\theta| \approx 100$ K, large compared to T_M . In spite of these interactions completely different from the RKKY ones, the low

temperature and field behaviour of AC susceptibilities, magnetizations ($M_{rs}(0) \approx 10^{-2} M_0$) are rather similar to those of usual spin-glass alloys. The spin-glass behaviour of such interesting amorphous materials could be due to frustration effects.

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