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CONCENTRATION DEPENDENCE OF CRYSTAL-FIELD EFFECTS IN AMORPHOUS $\text{Ce}_x \text{Si}_{1-x}$ ALLOYS

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Résumé - Les effets de champ cristallin sur les ions de cérium dans les alliages amorphes $\text{Ce}_x \text{Si}_{1-x}$ ($0.06 \leq x \leq 0.70$) sont analysés à travers la dépendance en température de la susceptibilité magnétique. La constante de Curie-Weiss déterminée à basse température présente une dépendance en concentration très marquée et passe par un minimum aux alentours de $x = 0.28$ suggérant un changement assez abrupt de la symétrie locale pour cette composition.

Abstract - Crystal-field effects on Ce ions in amorphous $\text{Ce}_x \text{Si}_{1-x}$ alloys ($0.06 \leq x \leq 0.70$) are analyzed through the temperature dependence of the magnetic susceptibility. The low-temperature Curie-Weiss constant exhibits a strong concentration dependence on the Si rich side to undergo a sharp minimum around $x = 0.28$, suggesting an abrupt change of the local symmetry about the Ce ions for this composition.

I - INTRODUCTION

Bulk magnetic properties of amorphous alloys have been shown to be sensitive in many cases to the atomic-scale arrangement /1/. In particular, the low-temperature magnetization together with the temperature dependence of the magnetic susceptibility in paramagnetic alloys containing rare earths (RE) elements can be strongly affected by the crystal-field (CF) energy level scheme. In the case of heavy RE with large J values, the CF effects can be approximated by the random uniaxial anisotropy model /2/. But, for RE with small J values, the non-axial components of the CF quadratic Hamiltonian have to be taken into account /3/. In this latter case, it is possible to gain some insight about the local symmetry about the RE ions in an amorphous matrix through the analysis of the bulk magnetization data /4/.

From the analysis of the low-temperature magnetization and of the temperature dependence of the susceptibility, one can obtain in principle a rough characterization of the CF energy level scheme for a RE ion. Among the elements of information that can be readily gained are namely :

- i) the nature of the ground state characterized by its saturation moment and its effective moment,
- ii) an estimate of the energy gap between the ground state and the first excited states,
- iii) an estimate of the overall CF splitting.

From such information, the local symmetry itself cannot be readily determined. But by following the concentration dependence of these parameters, one can expect to detect in a family of amorphous alloys any change in local symmetry as a function of composition.

We report here on a study of CF effects in amorphous $\text{Ce}_x \text{Si}_{1-x}$ alloys ($0.06 \leq x \leq 0.70$) through bulk magnetization measurements. These alloys are suitable for such an investigation for the following reasons :

- i) the Ce ions were shown by X-ray absorption edge measurements to be purely trivalent within the whole concentration range in the amorphous state /5/,
- ii) the magnetic ordering temperatures are low enough for the exchange effects to be negligible above 20 K, as will be discussed along later.

II - EXPERIMENTAL

Amorphous $\text{Ce}_x\text{Si}_{1-x}$ alloys were produced by co-evaporation onto liquid N_2 cooled substrates under ultra-high vacuum. The amorphous nature of the films was ensured by electron microscopy. Other details concerning the samples can be found in a previous report /6/.

Magnetic measurements were performed between 1.6 and 400 K in maximal field 50 kG. Appropriate corrections were made for the substrate and for the protecting Si film.

III - RESULTS AND DISCUSSION

In good agreement with our X-ray absorption edge measurements, no trace of valence admixture for Ce was found in our magnetic data above 1.6 K in all the amorphous $\text{Ce}_x\text{Si}_{1-x}$ alloys we investigated, including within the concentration region where the crystalline CeSi_2 compound exhibits a Kondo-lattice type behaviour /7/. The trivalent character of Ce ions in amorphous CeSi_2 at variance with the non-magnetic behaviour of Ce in crystalline CeSi_2 is illustrated on fig. 1 by the magnetization versus field at 4.2 K.

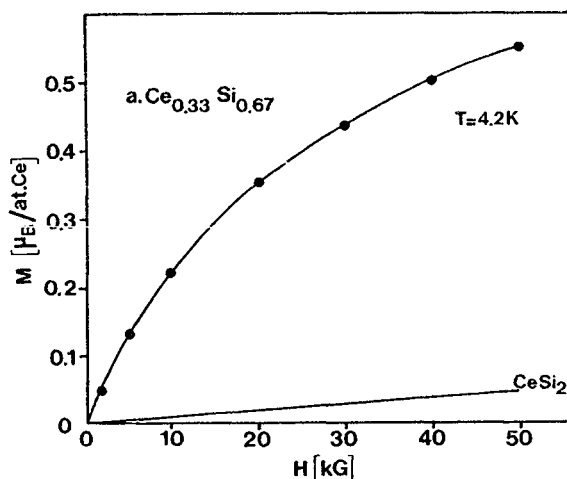


Fig. 1 - Magnetization versus field at 4.2 K in amorphous and crystalline CeSi_2 (Data for crystalline CeSi_2 are after ref. /7/).

A typical experimental curve for the temperature dependence of the initial susceptibility is shown on fig. 2. This curve can be divided into three parts. The *low-temperature* part for alloys with $x \geq 0.35$ is dominated by exchange effects. Indeed, these alloys undergo a low-temperature spin-glass type of transition, as illustrated on fig. 3. The origin of the low-temperature upturn of the susceptibility for more dilute alloys is not clearly identified. It might be due to some other RE impurities in the cerium. The *high-temperature* part of the susceptibility progressively approaches the ionic value, but without reaching it even at 400 K. This clearly indicates that the CF overall splitting in this family of alloys is anomalously high. Finally, the temperature dependence of the susceptibility within the *medium-temperature* range is of the Curie-Weiss type, yielding a Curie-Weiss constant which characterizes the ground state.

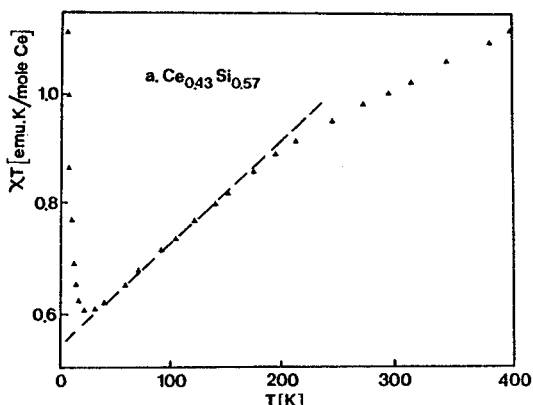


Fig. 2 - Initial susceptibility ($H \leq 1$ KG) multiplied by temperature versus temperature for amorphous $\text{Ce}_{0.43}\text{Si}_{0.57}$. (The ionic Curie-Weiss constant for Ce^{3+} is 0.806 emu/mole Ce).

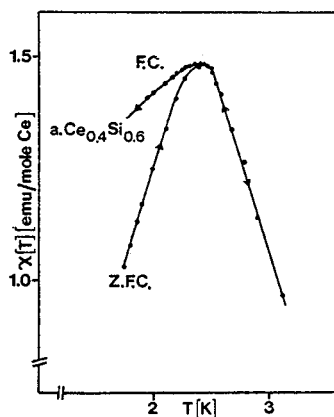


Fig. 3 - Initial susceptibility versus temperature for amorphous $\text{Ce}_{0.4}\text{Si}_{0.6}$. The field cooled ($H = 30$ Oe) curve departs from the zero-field cooled curve at the temperature of the susceptibility maximum.

On fig. 4, we plotted the low-temperature Curie-Weiss constant C as a function of cerium content. Above $x \approx 0.40$, C decreases smoothly when Ce content increases. Below $x \approx 0.10$, C is practically constant. A very sharp reduction of C is observed between $x = 0.10$ and $x = 0.40$, with a sharp minimum for $x \approx 0.28$. As expected, a similar concentration effect can be also detected on the low-temperature magnetization (fig. 5). A Curie-Weiss law holds. This temperature range is about 100 K for the alloys with $x \geq 0.40$; it decreases down to 50 K for $x = 0.28$ to remain constant and equal to 50 K for more dilute alloys. As stated above, such a temperature range yields an indication on the energy gap between the ground state and the first excited states. This gap is found to be large, in agreement with the high value speculated for the CF overall splitting. On the other hand, the change for this gap observed around $x = 0.28$ together with the minimum for the low-temperature Curie-Weiss constant suggest a change in the local symmetry occurring for this composition. It can be also noted that an extrapolation of the temperature for the spin-glass transition versus Ce content (fig. 6) yields a concentration close to $x = 0.30$.

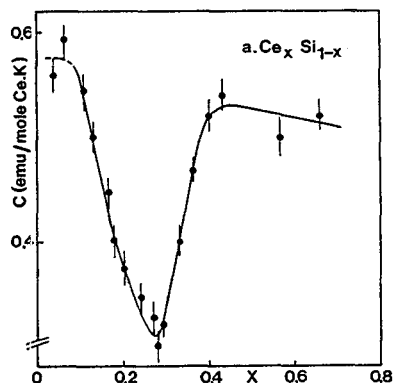


Fig. 4 - Low-temperature Curie-Weiss constant C as a function of Ce content in amorphous $\text{Ce}_x \text{Si}_{1-x}$ alloys.

EXAFS measurements on Ce in $\text{a.Ce}_{0.28}\text{Si}_{0.72}$ and on alloys on both sides of this composition are in progress in order to characterize the change in the local order around $x = 0.28$. This change is not likely to be related with the metal-non metal transition occurring around $x = 0.15$ according to our resistivity measurements. More likely, this change could be related with the local order of the crystalline CeSi_2 compound, the stoichiometry of which is under investigation (K. A. GSCHNEIDNER, private communication)

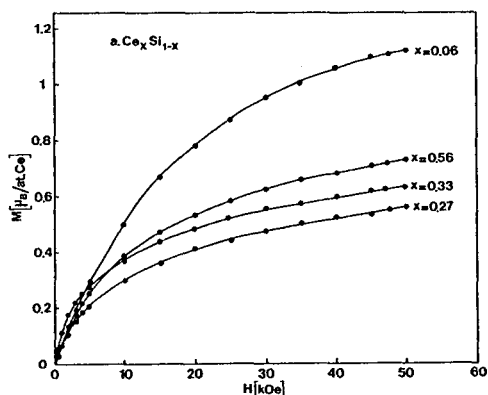


Fig. 5 - Magnetization versus field at 1.7 K for several amorphous $\text{Ce}_x \text{Si}_{1-x}$ alloys.

Finally let us comment on the values we obtained for the low-temperature Curie-Weiss constant. In any case, our values for C are not compatible with the random uniaxial assumption ($C = 0.575$ emu/mole Ce). Values for dilute Ce alloys and for concentrated $\text{Ce}_x \text{Si}_{1-x}$ alloys can be accounted for by a CF quadratic Hamiltonian ($0.438 \leq C \leq 0.575$ emu/mole Ce). Low values obtained for $0.20 \leq x \leq 0.30$ alloys are not compatible with a purely quadratic Hamiltonian, suggesting a richer symmetry. Similar departures from the quadratic model were already observed in amorphous $\text{Ce}_{72} \text{Cu}_{28}$ and $\text{Ce}_{89} \text{Al}_{11}$ alloys [8/.

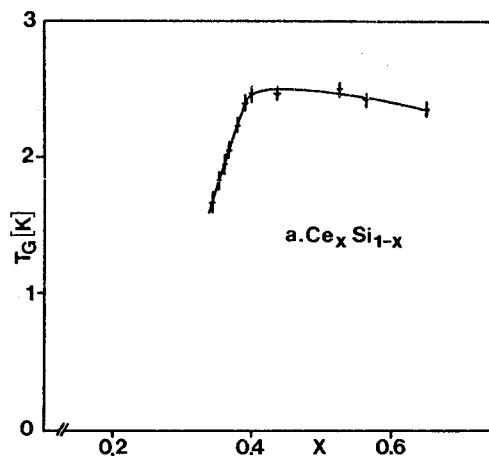


Fig. 6 - Temperature of the susceptibility maximum (T_G) versus Ce content in amorphous $\text{Ce}_x \text{Si}_{1-x}$ alloys.

IV - CONCLUSION

A study of CF effects through bulk magnetic measurements shows evidence for a drastic change in local symmetry occurring around $x = 0.28$ in amorphous $\text{Ce}_x \text{Si}_{1-x}$ alloys. Further local measurements are clearly needed to properly characterize this change. Meanwhile, this clearly illustrates the ability of magnetic measurements as a tool of investigation for the local order in amorphous alloys beyond the RDF information obtained by standard diffraction techniques.

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