Proceedings of the 15th Czech and Slovak Conference on Magnetism, Košice, Slovakia, June 17–21 2013

Magnetic Properties of $Ce(Cu_xNi_{1-x})_4Mn$ Compounds

K. Synoradzki*, T. Toliński
Institute of Molecular Physics, Polish Academy of Sciences, Smoluchowskiego 17, 60-179 Poznań, Poland

The $Ce(Cu_xNi_{1-x})_4Mn$ series of compounds has been studied for a wide range of the x values. The magnetic properties are determined from the temperature dependence of the dc and ac magnetization. A large irreversibility is observed between the zero-field cooled (ZFC) and field-cooled (FC) curves with a maximum in the ZFC one. The ac magnetic susceptibility also shows a peak, which shifts to higher temperatures with increasing frequencies, which is typically observed in spin-glass (SG) systems. The observation of the SG behaviour is further supported by a very slow decay of the isothermal remanent magnetization and by the typical shape of the magnetization curves. A range of x values showing the para-ferromagnetic-SG transitions has been found.

DOI: 10.12693/APhysPolA.126.300
PACS: 75.50.Lk, 71.20.Lp

1. Introduction

The $CeNi_4Mn$ compound is a ferromagnet with $T_C =$ 130 K, whereas $CeCu_4Mn$ is a spin-glass system. In the previous study [1, 2] we have shown for $CeCuNi_3Mn$ that the Cu substitution develops a spin-glass (SG) state. To further investigate this SG behaviour we have now prepared a number of samples in the series $Ce(Cu_xNi_{1-x})_4Mn$. This study of an isostructural series of compounds is directed towards the question of the stability of the spin-glass state against variations of the Cu concentration.

2. Experimental details

The intermetallic compounds $Ce(Cu_xNi_{1-x})_4Mn$ were prepared from the pure elements in argon atmosphere inside a water-cooled sample chamber of an induction furnace. The powder X-ray diffraction technique (Co-K_α radiation) confirmed that all the prepared compounds crystallize in the hexagonal $CaCu_5$-type structure (space group P6/mmm).

The lattice parameters were acquired from the diffraction patterns using the FullProf program and they obey the Vegard's law.

The dc and ac magnetic susceptibility were measured in the temperature range 1.9–300 K using the PPMS (Quantum Design) commercial device.

3. Results

Figure 1 shows the x-ray diffraction patterns along with the fitting curves. The broad peaks for the $Ce(Cu_xNi_{1-x})_4Mn$ compounds indicate that the chemical inhomogeneity becomes rapidly significant after the Cu substitution. This can induce magnetic inhomogeneity and influence the magnetic behaviour of the samples. The substitution of Ni by Cu atoms affects significantly the lattice parameters and the cell volume. The crystal lattice parameters and the unit cell volume increase linearly as the Cu content increases. A lack of deviation from the Vegard's rule may indicate that the cerium atoms are present as stable trivalent ions in the $Ce(Cu_xNi_{1-x})_4Mn$ compounds.

The temperature dependent zero field cooled (ZFC) and field cooled (FC) dc magnetization M_{dc} in magnetic field of 1 kOe is plotted for exemplary samples in Fig. 2. The strong irreversibility between the ZFC and FC magnetization curves, typical of spin-glass like systems is observed. This irreversibility decreases with increasing the applied magnetic field (see Fig. 2a, inset). The high-temperature ($T > 140$ K) susceptibility data of all samples can be very well fitted with the Curie-Weiss law.

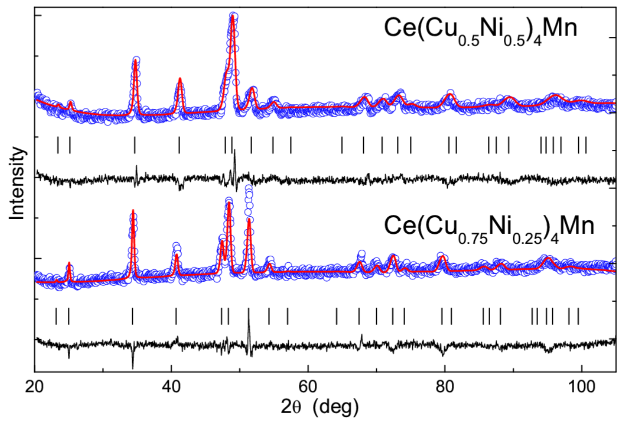

Fig. 1. X-ray diffraction patterns along with the fitting curves for the $Ce(Cu_xNi_{1-x})_4Mn$ ($x = 0.5$ and 0.75) samples.

For several samples ($x = 0.375$, 0.5, 0.625 and 0.75) the magnetization data shows a sharp onset of the magnetic order at $T_C = 79$, 115, 123 and 78 K, respectively. The values of the temperatures of the ferromagnetic (FM) to paramagnetic (PM) phase transition (T_C) were extracted from the $M_{FC}(T)$ curves as the inflection point indicated by the minimum of dM/dT (Fig. 2c, inset). The freezing temperatures T_f were obtained as a position of the maximum of the ZFC curve. Therefore the $M - T$ data is indicative of a possible SG phase below the Curie tem-

*corresponding author; e-mail: karol.synoradzki@ifmpan.poznan.pl

perature in these compounds. Figure 3 shows a magnetic phase diagram for the Ce(Cu$_x$Ni$_{1-x}$)$_4$Mn system, where T_f and T_C were obtained from the $M_{dc}(T)$ data. T_f changes almost linearly, which is often observed in spin glass systems.

The appearance of the glassy state in the vicinity of the FM transition gives rise to two possible types of the SG behaviour: reentrant SG (RSG) or cluster glass (CG). RSG systems undergo PM to FM ordering at T_C and the SG transition takes place much below the FM transition, developing a plateau-like feature in the ZFC and FC curves. On the other hand the CG state can be considered as a set of clusters formed around T_C due to a short range FM ordering. The freezing of the assembly of clusters takes place at a low temperature by overcoming the energy barrier induced by the local anisotropy [3].

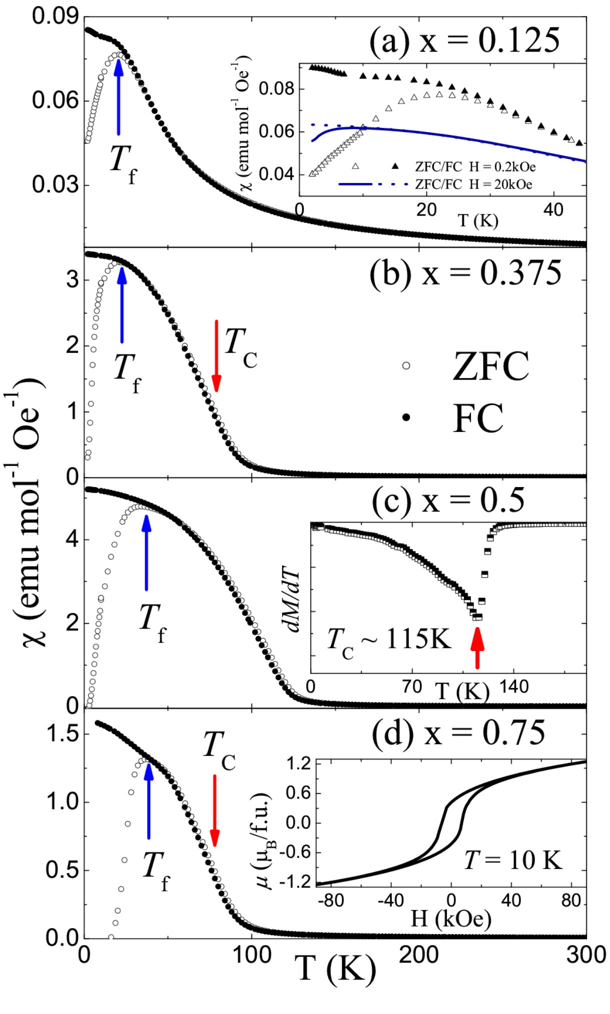

Fig. 2. Temperature variation of the dc magnetization of Ce(Cu$_x$Ni$_{1-x}$)$_4$Mn ($x = 0.125$, 0.375, 0.5 and 0.75) compounds in ZFC and FC modes at $H = 1$ kOe. Insets: see the text.

At low temperatures $M(H)$ loops (Fig. 2d, inset) show a pronounced S-shaped curves that are characteristic of SG, which dissolve into linear curves with increasing temperature. For intermediate x values the S-shaped curves exhibit a hysteresis and coercivity at low fields, indicating a coexistence of the SG and FM states. Also, it can be observed that even under the highest magnetic field of 90 kOe the magnetization remains unsaturated, indicating that no canonical long range order exists in the Ce(Cu$_x$Ni$_{1-x}$)$_4$Mn system. This suggests that the formation of clusters is due to a short range order and results in the CG state in these compounds [3].

To confirm the SG state, the temperature dependences of the real χ' and imaginary χ'' components of the magnetic susceptibility were measured at different fixed frequencies ranging from 10 Hz to 10 kHz. For samples with $x = 0.375$, 0.5, 0.625 and 0.75, a double-anomaly was observed (not shown). The higher temperature inflexion corresponds to PM-FM transitions and the frequency dependence of the peak at T_C is found to be quite negligible for both the in-phase and the out-of-phase components. The second anomaly is shifted to higher temperatures with increasing the frequency f, which is characteristic of the SG state. Moreover, the dependence $T_f(f)$ obeys the Vogel-Fulcher law and the conventional critical slowing down model.

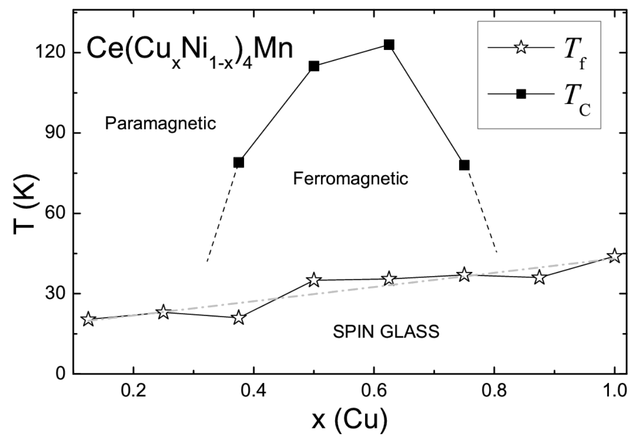

Fig. 3. The magnetic phase diagram for Ce(Cu$_x$Ni$_{1-x}$)$_4$Mn. T_f and T_C were obtained from the $M_{dc}(T)$ data.

4. Conclusions

The magnetic properties of the Ce(Cu$_x$Ni$_{1-x}$)$_4$Mn system have been investigated. Dc and ac magnetic susceptibility measurements have revealed that the samples with $0.375 \le x \le 0.75$ undergo the PM to FM transition, followed by the SG transition, while the samples with $x = 0.125$, 0.25 and 0.875 show only the SG behaviour. It seems that the system does not possess a canonical SG state, rather a CG state is observed, which bears similar features to those of SG and RSG.

References

[1] T. Toliński, K. Synoradzki, *Intermetallics* **19**, 62 (2011).

[2] K. Synoradzki, T. Toliński, *Acta Phys. Polonica A* **121**, 1079 (2012).

[3] R. Nigam, A.V. Pan, S.X. Dou, *J. Appl. Phys.* **105**, 07E303 (2009).