

THERMAL TREATMENT EFFECTS
ON THE SPIN-GLASS-LIKE STATE
OF THE $\text{CeNi}_{0.8}\text{Cu}_{0.2}$ ALLOY*

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We have investigated the modifications induced by thermal treatments in the structural and magnetic properties of the $\text{CeNi}_{0.8}\text{Cu}_{0.2}$ sample which presents a spin-glass-like behaviour below 6 K. No remarkable changes in the microstructure have been detected by X ray diffraction and scanning electron microscopy. However, the magnetic properties depend on the thermal treatment of the sample. This fact suggests that modifications in the size and number of the magnetic clusters are being induced. In addition, long-range magnetic order is evidenced below 0.6 K.

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The orthorhombic pseudobinary $\text{CeNi}_{0.8}\text{Cu}_{0.2}$ compound does not present long range magnetic order down to 2 K. However, ac susceptibility (χ_{ac}) measurements reveal a spin-glass-like state below 6 K [1], characterised by a maximum at that temperature. This anomaly is observed in all the “as quenched” prepared samples of this composition. Further ac-dc magnetisation analysis [2], together with μSR studies [3], suggest the existence of

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an inhomogeneous spin-glass consisting on magnetic clusters. In this cluster glass state, aggregates of magnetic ions would be embedded in a mostly non-magnetic matrix. Hence, this state should be very sensitive to different thermal treatments. In order to test this picture we have investigated these effects on the structural and low temperature properties of different preparations of the alloy $\text{CeNi}_{0.8}\text{Cu}_{0.2}$.

The as quenched (AQ) $\text{CeNi}_{0.8}\text{Cu}_{0.2}$ sample used in references [1,2] is a polycrystal prepared by arc melting stoichiometric amounts of high purity elemental metals under an Ar atmosphere. A piece of this sample was annealed (AN) at 440°C for one week. After characterising the AN sample, it was re-melted (RM) under the same conditions as the AQ sample.

Crystalline and metallurgical characterisation have been done by X Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM). Magnetic ac-dc measurements down to 300 mK were carried out using a SQUID magnetometer at the CRTBT (Grenoble).

In Fig. 1 the XRD patterns of the AN and RM samples at room temperature are presented. Both are characteristic of the FeB-type of structure (Pnma space group). Spurious phases were almost negligible (less than 2%). The calculated patterns (solid lines in Fig. 1) have been obtained placing the Ce ions in a $4c$ site and the Cu/Ni randomly distributed on the other $4c$ site. To our surprise, we have to highlight that the annealing procedure does not significantly improve the crystallinity of the sample, as no mean-

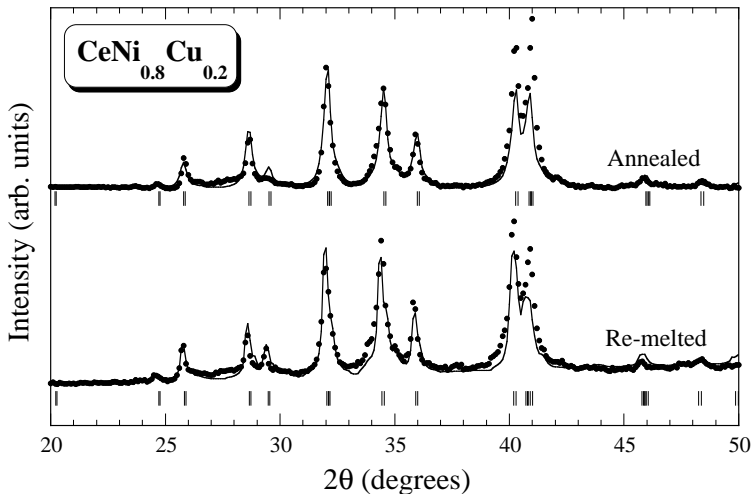


Fig. 1. The observed (dots) and calculated (lines) X ray powder diffraction patterns of the AN and RM samples. The vertical ticks at the bottom of each plot indicate the calculated Bragg peak positions.

ingful changes are observed in the peaks width. On the other hand, a slight decrease of the cell parameters is observed (less than 1%).

With respect to the SEM results, the intensity of backscattered electron images yields semiquantitative information about composition homogeneity in a given sample. This information can be improved performing a quantitative electron probe microanalysis. We also used secondary electron images to distinguish composition fluctuations from physical voids in the sample.

The images reveal that both the AN and RM samples mainly consist of a matrix of single phase material. Furthermore, the electron probe microanalysis indicates that this matrix presents the proper composition and is homogeneous down to the available magnification for this kind of analysis (several μm). Then, if the annealing process induces any change on this sample, it has to take place at a smaller scale (nanometric).

In Fig. 2, the low temperature χ_{ac} is compared for the AN and RM samples, also showing its frequency dependency by plotting three selected measurements at $\nu = 2.11$, 21.1 and 211 Hz for each sample. The main effect

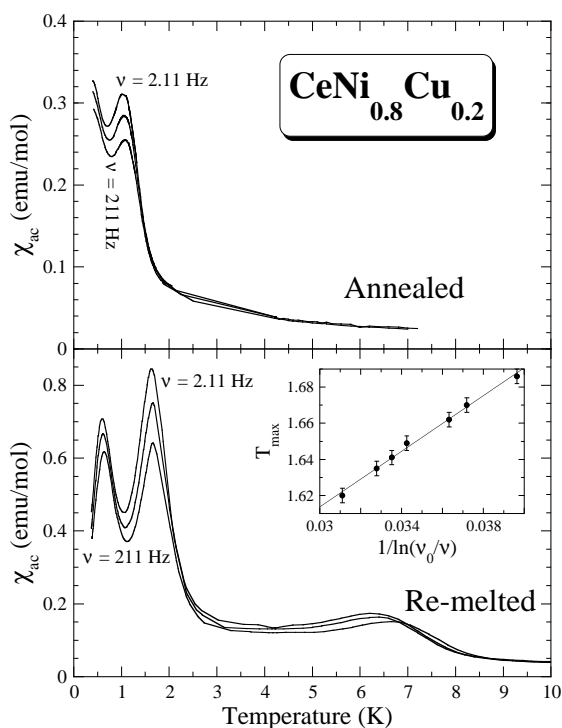


Fig. 2. Low temperature evolution of χ_{ac} at three selected frequencies ($\nu = 2.11$, 21.1 and 211 Hz) for the AN and RM samples. The inset shows the shift with frequency of the temperature maximum found at 1.6 K fitted to a Vogel–Fulcher law.

of the annealing is to annihilate the maximum observed at 6 K for the AQ samples, which was related with the transition into a spin-glass-like state. In addition, another low temperature anomaly is detected around 1 K. The frequency shift of this maximum is $\delta = (\Delta T_{\max}) / (T_{\max} \Delta \log \nu) = 0.023$.

The RM sample recovers the transition at 6 K and two additional maxima appear at low temperatures: 1.6 and 0.6 K. From our analysis it is inferred that these maxima correspond to transitions already present in the AN sample that have shifted to higher temperatures. In fact, the maximum at 1.6 K also exhibits a frequency shift ($\delta = 0.014$) of the same order of magnitude as that found for the AN sample, and certainly, the lowest transition was not seen for that sample due to the experimental limitations.

The already reported δ values are characteristic of spin-glass systems [4, 5]. However, the lowest temperature maximum seems to be related with a transition into a long-range ordered state, even for the AN sample. This suggestion is strongly supported by the Zero Field and Field Cooled magnetisation measurements and by the $M(H)$ curves obtained at different temperatures (not shown by lack of space). The existence of a long-range ordered state at low temperatures for both AN and RM samples has also been confirmed by recent μ SR studies [6].

The absence of the spin-glass transition at 6 K in the AN sample reasserts the assumption of a cluster spin-glass state done for the previously studied AQ samples. Although the structural studies do not indicate qualitative differences in the two different preparations, clear modifications in the magnetic properties with the thermal treatment have been found. Thus, the annealing has significantly altered the size and spatial distribution of the clusters. All these results evidence the existence of inhomogeneities (clusters) at the nanoscopic scale that are in the basis of the complex magnetic behaviour of this system.

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