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Multi-magnetic phases in $Fe_{1-x}Ni_xTa_2O_6$

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Abstract

X-ray, neutron diffraction and magnetic susceptibility are reported for $Fe_xNi_{1-x}Ta_2O_6$ mixed oxides. X-ray refinement indicates homogeneous samples for all the reported concentrations. The neutron-diffraction measurements reveal magnetic structures with double propagation vectors. This system exhibits at least two bicritical points at about $x = 0.15$ and 0.60. For these concentrations, at low temperatures, the system shows the coexistence of two magnetic structures. This bicritical behaviour is interpreted as induced by competition between the different magnetic structures.

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Keywords: Neutron diffraction; Magnetic phases coexistence

1. Introduction

Pseudoternary oxides $(A_xB_{1-x})Ta_2O_6$, for A, B = Fe, Co and Ni, crystallize in the trirutile structure in space group $P4_2/mnm$ for all intervals of concentration. The 3d transition-metal ions have the same symmetry as that of Ni in the antiferromagnet K_2NiF_4, well characterized by short-range two-dimensional correlations followed by a crossover to a three-dimensional long-range order at low temperature [1]. The susceptibility curve shows a broad maximum and falls off slowly in the paramagnetic region. Below this maximum a subtle inflection is observed [2–4]. The broad maximum is indicative of short-range correlations, while the inflection point is associated to the long-range order, as indicated by specific heat measurements.

Magnetic susceptibility measurements show a Néel temperature, T_N, of about 9.5 K for $FeTa_2O_6$ and 7.1 K for $CoTa_2O_6$ [5]. The magnetic structures for these compounds were found by neutron diffraction and could be indexed with double propagation vectors: (1/2, 0, 1/2) and (0, 1/2, 1/2) for $FeTa_2O_6$ and ($\pm$1/4, 1/4, 1/4) for $CoTa_2O_6$ [5]. For $NiTa_2O_6$, specific-heat measurements give $T_N = 10.5$ K and the neutron-diffraction pattern is indexed with ($\mp$1/4, 1/4, 1/2) [6,7]. The T vs. x phase diagram for the system $Fe_xCo_{1-x}Ta_2O_6$ exhibits a bicritical point at about $T = 4.9$ K and $x = 0.46$ [5]. That is to say, at this Fe concentration and lower temperatures the system shows coexistence of both magnetic structures. This novel bicritical behaviour is interpreted as induced by competition between the different magnetic structures and crystallographic changes.

In this paper, we report crystallographic and magnetic structures of the system $Fe_xNi_{1-x}Ta_2O_6$.

2. Experimental

The synthesis procedure was the same as for $Fe_xCo_{1-x}Ta_2O_6$ [8,9], but heated at 1600 K. X-ray diffraction (XRD) was performed with a Siemens diffractometer

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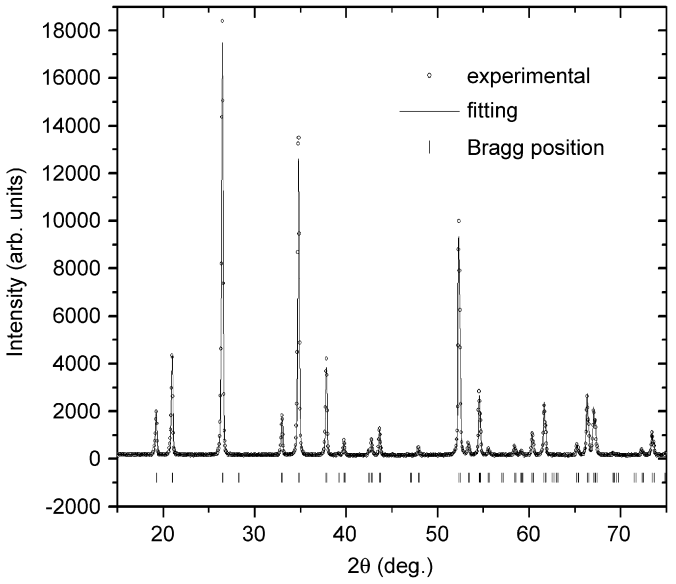

Fig. 1. Room temperature X-ray powder diffraction pattern for $Fe_{0.85}Ni_{0.15}Ta_2O_6$. The points represent the experimental data and the solid line represents the calculated pattern with the Rietveld refinement. Vertical tics are the $P4_2/mnm$ reflections.

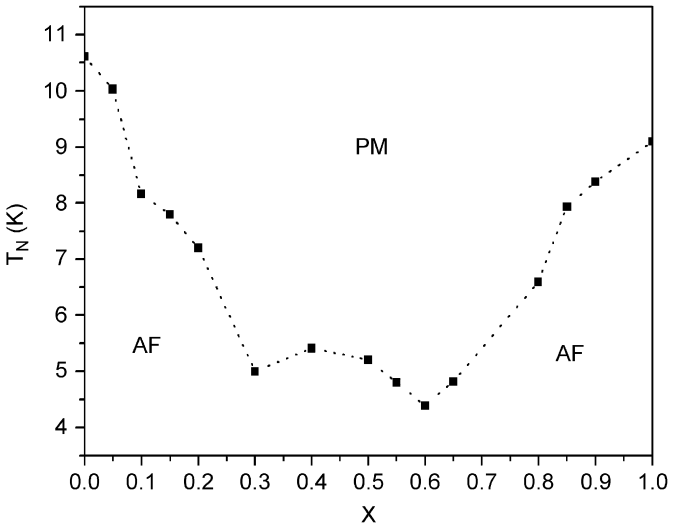

Fig. 2. T vs. x phase diagram, showing the paramagnetic (PM) phase and the region of antiferromagnetic ordering (AF). Solid points are the T_N obtained from magnetic susceptibility measurements. Broken lines are guides to the eyes.

D500 using CuKα radiation. Magnetic susceptibility was carried out with a SQUID magnetometer, from 3.5 to 300 K, in a 1 kOe applied field. Neutron powder-diffraction (ND) experiments were made at the ILL (CRG-D1B diffractometer; $\lambda = 2.52$ Å), Grenoble, France. Crystal and magnetic structures were refined using the FULLPROF program [10].

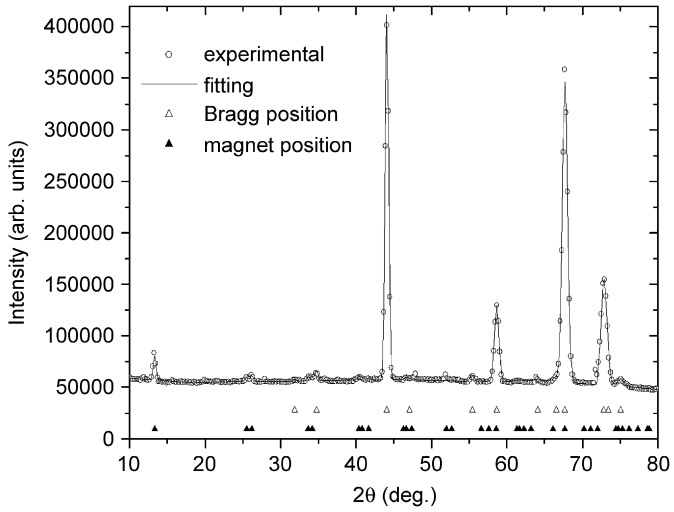

Fig. 3. Neutron-diffraction pattern for $Fe_{0.05}Ni_{0.95}Ta_2O_6$ at 1.8 K. Open symbols are the $P4_2/mnm$ reflections and solid ones the magnetic reflections indexed by propagation vectors ($\pm 1/4$, 1/4, 1/2).

3. Results

All the samples are tetragonal and have been indexed to the space group $P4_2/mnm$, as expected. A typical pattern is displayed in Fig. 1 for the sample $Fe_{0.85}Ni_{0.15}Ta_2O_6$.

Magnetic susceptibility curves (not shown) exhibited a broadened maximum typical of two-dimensional systems. The long-range order temperature T_N was obtained from the maximum in the curves $\partial(\chi T)/\partial T$ vs. T. Fig. 2 shows the T_N vs. x plot. The minimum around $x = 0.60$, as in the case of $Fe_xCo_{1-x}Ta_2O_6$ [5], is associated to a bicritical point with a coexistence of magnetic phases.

A typical neutron-diffraction pattern at 1.8 K is shown in Fig. 3 for $Fe_{0.05}Ni_{0.95}Ta_2O_6$. For temperatures below T_N all the magnetic structures are indexed with double propagation vectors: ($\mp 1/4$, 1/4, 1/2) for $NiTa_2O_6$ and (1/2, 0, 1/2) + (0, 1/2, 1/2) for $FeTa_2O_6$. The latter remain unchanged in the Fe-rich samples ($0.60 \leqslant x \leqslant 1.00$), while the propagation vectors are ($\mp 1/4$, 1/4, 1/2) for the Ni-rich samples ($0.00 \leqslant x \leqslant 0.15$). For $x = 0.15$, the system shows the coexistence of two magnetic structures with propagation vectors ($\mp 1/4$, 1/4, 1/2) and ($\mp 1/4$, 1/4, 0). The same happens for $x = 0.60$, but with vectors ($\pm 1/4$, 1/4, 1/4) and (1/2, 0, 1/2) + (0, 1/2, 1/2). In Fig. 4 we observe the evolution of magnetic phases with Fe concentration, where we detected four different magnetic structures and the coexistences at $x = 0.15$ and 0.60.

In conclusion, we have shown that the antiferromagnetic system $Fe_xNi_{1-x}Ta_2O_6$ exhibits four magnetic structures at temperatures below T_N with at least two regions of coexistence of phases.

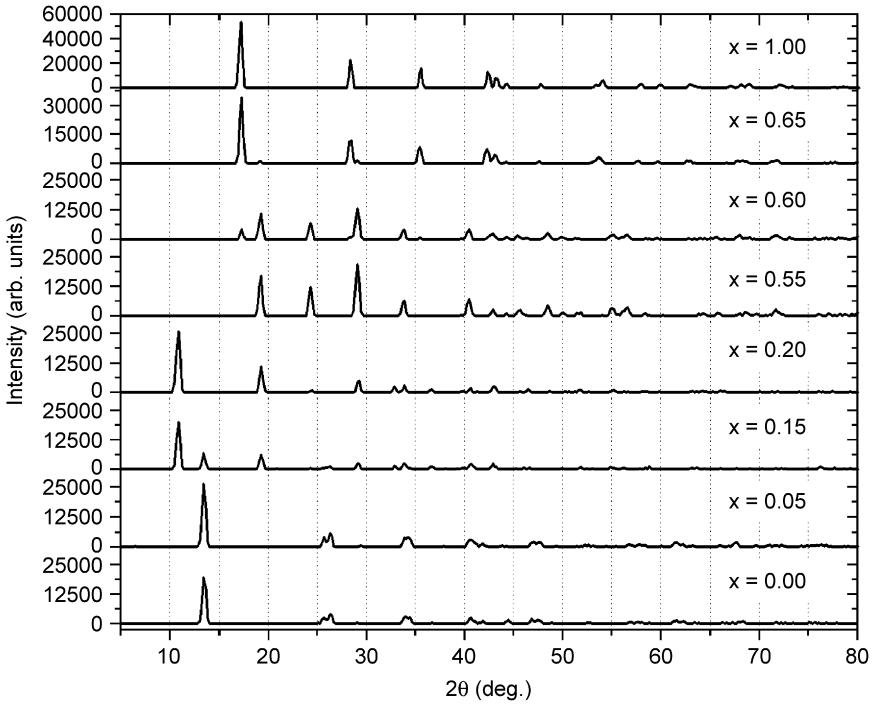

Fig. 4. Neutron-diffraction pattern of the magnetic structure for $Fe_xNi_{1-x}Ta_2O_6$. The crystallographic contribution was subtracted.

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