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luizno.bjp@gmail.com

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# Magnetic, Structural and Superconducting Properties of $\text{MgC}(\text{Ni}_{1-x}\text{Fe}_x)_3$

M. Alzamora, D. R. Sánchez, M. Cindra, and E. M. Baggio-Saitovitch

*Centro Brasileiro de Pesquisas Físicas,*

*Rua Xavier Sigaud 150, Urca, CEP 22290-180 Rio de Janeiro, Brasil*

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X-ray diffraction, ac susceptibility and  $^{57}\text{Fe}$  Mössbauer spectroscopy measurements have been performed in the  $\text{MgC}(\text{Ni}_{1-x}\text{Fe}_x)_3$  ( $0 \leq x \leq 0.5$ ) series. The superconducting transition temperature  $T_c$  of the undoped sample, determined by ac susceptibility, is  $\sim 7\text{K}$ . The Fe doping quickly depresses  $T_c$ , completely destroying the superconducting state for  $x \sim 0.04$ . The establishment of an ordered magnetic state is only observed for Fe concentration ( $x \geq 0.3$ ) far above the concentration for which the superconducting state has completely disappeared. An expansion of the lattice with the increase of the Fe content was observed.

## I Introduction

The discovering of superconductivity in the  $\text{MgB}_2$  compound with  $T_c \sim 39\text{K}$  [1] has renewed the interest for the search of new intermetallic compounds with high superconducting transition temperatures. Recently, another intermetallic compound containing Mg was discovered:  $\text{MgCNi}_3$ , with  $T_c \sim 8\text{K}$  [2]. Different of the layered  $\text{MgB}_2$  (space group  $P6/mmm$ ), where the boron layers are alternately stacked on the Mg layers, the  $\text{MgCNi}_3$  has the classical cubic perovskite structure ( $Pm\bar{3}m$ ), resembling the structure of the high  $T_c$  superconductors. However, in contrast to the high  $T_c$  perovskites, the  $\text{MgCNi}_3$  has no oxygen in its structure. Furthermore, the establishment of a superconducting than a ferromagnetic ground state in a intermetallic compound with a high Ni content (element usually associated to ferromagnetism) is very surprising and make it a potential candidate for unconventional superconductivity, as pointed out by T. He [2].

The superconducting properties of the  $\text{MgCNi}_3$  are associated with the occurrence of an intense peak in the density of Ni  $3d$  states at the Fermi level. In fact, the Fermi level is located in the slope descending of this peak [3-7]. In the same way, these calculations suggest that introducing electrons or hole dopants into  $\text{MgCNi}_3$  will result in a decrease or increase in  $N(E_F)$ , respectively. In the former case (substituting Ni by Cu), one may expect that superconductivity of the system deteriorate. Doping with holes (e.g. Ni by Fe) will increase the  $N(E_F)$ . However, a large  $N(E_F)$  is not always beneficial for superconductivity, because a very large  $N(E_F)$  tends to induce spin fluctuation or mag-

netic order, which quickly destroy the superconducting electron pairing.

The influence of Ni substitution by Fe, on the structural, magnetic and superconducting properties of the  $\text{MgC}(\text{Ni}_{1-x}\text{Fe}_x)_3$  ( $0 \leq x \leq 0.5$ ) system has been investigated, in this work, by x-ray, magnetization and Mössbauer spectroscopy of  $^{57}\text{Fe}$ .

## II Experimental

Samples with nominal formula  $\text{MgC}(\text{Ni}_{1-x}\text{Fe}_x)_3$  for  $0 \leq x \leq 0.5$  where prepared and the starting materials were bright Mg flakes, fine Ni powder, and high purity amorphous carbon powder. The reactions were done in a tube furnace under high-pure Ar atmosphere for half an hour at  $600^\circ\text{C}$ , followed by an hour at  $900^\circ\text{C}$ . After cooling, the samples were grained again, pressed into pellets, and sintered for more one hour at  $900^\circ\text{C}$ . The structural characterization were made by powder X-ray diffraction using a Rigaku X-ray diffractometer with Cu  $K_\alpha$  radiation. The samples were analyzed by Mössbauer spectroscopy in transmission geometry in the temperature range of 2-300K. AC susceptibility measurements were performed in a Quantum Design SQUID magnetometer.

## III Results and Discussion

To obtain stoichiometric composition  $\text{MgC}(\text{Ni}_{1-x}\text{Fe}_x)_3$ , excess of carbon and magnesium were required [2,8,9].

The X-ray spectra (Fig. 1a) for all the samples show the single cubic  $\text{MgCNi}_3$  ( $Pm3m$ ) as a majority phase with a small amount of unreacted graphite. After Rietveld

analysis, an expansion of the lattice with the increase of the Fe doping is determined (Fig. 1b).

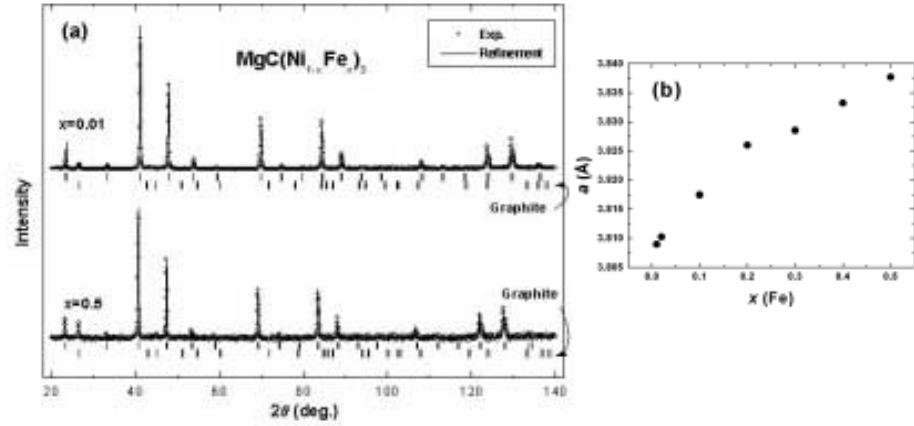


Figure 1. (a). Cu  $K\alpha$  x-ray diffraction pattern of the samples  $\text{MgC}(\text{Ni}_{1-x}\text{Fe}_x)_3$  with  $x=0.01$  and  $x=0.5$ . The Bragg peaks of the remaining graphite are indicated. (b) Lattice parameter  $a$  as a function of Fe content.

The ac susceptibility measurements, showing the superconducting and magnetic transition of the doped materials, are displayed in Fig 2a. The  $T_c \sim 7\text{K}$  of the undoped sample is in agreement with previous data obtained from dc magnetization measurements [1,8]. The Fe dilution at the Ni site induces a fast decrease of the superconducting transition temperature from  $T_c \sim 7\text{ K}$  ( $x = 0.0$ ) to  $T_c \sim 3\text{ K}$  ( $x = 0.02$ ), and the disappearance of the superconducting state is extrapolated to occurs at  $x \sim 0.04$  (Fig. 2b). This suppression of superconductivity can, in principle, be explained considering band structure calculations: as mentioned before, for the parent  $\text{MgCNi}_3$  compound, the Fermi energy  $E_F$  is on the high-energy side of an intense peak of the density of

Ni  $3d$  states [3-7]. Doping the compound with Fe (hole doping) reduces the number of valence electrons and moves the Fermi level to lower energies, which causes an increase in the DOS at the Fermi level  $N(E_F)$ . According to the BCS theory,  $T_c$  should rise with the increase in  $N(E_F)$ , and not decrease as observed. However, an increase in the DOS leads to enhancement of the Stoner exchange parameter  $S=N(E_F)I_{ex}$  ( $I_{ex}$  is the exchange integral), and hence inducing spin fluctuations or magnetic order, which should destroy the superconducting pairing [4,5]. In the inset of Fig. 2 can be observed that a magnetic transition occurs only for Fe concentrations  $x \geq 0.3$ .

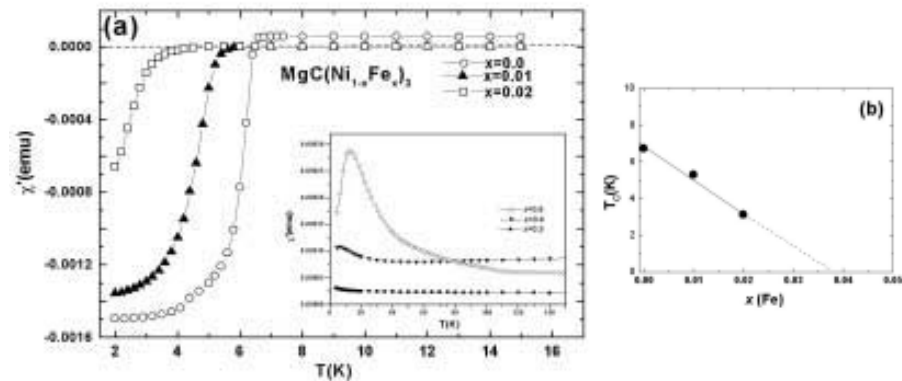


Figure 2. (a). Magnetic characterization of the superconducting and magnetic transitions of  $\text{MgC}(\text{Ni}_{1-x}\text{Fe}_x)_3$  ( $0 \leq x \leq 0.3$ ) samples. (b)  $x$ -dependence of the superconducting transition temperature of  $\text{MgC}(\text{Ni}_{1-x}\text{Fe}_x)_3$

$^{57}\text{Fe}$  Mössbauer experiments were performed in  $\text{MgC}(\text{Ni}_{1-x}\text{Fe}_x)_3$  system in order to reveal the referred magnetic fluctuations responsible, as pointed out by band filling, for the disappearance of the superconductivity in this series. At room temperature the spectra show a single quadrupole doublet (Fig. 3), which is attributed to Fe in the  $\text{MgCNi}_3$  structure. The sharp resonance lines indicate that there is a single well defined site for Fe in these compounds. In some samples an additional weak doublet (5% of total absorption area) was observed, which is probably due to impurity phase. The electrical quadrupole splitting  $\Delta E_Q$ , a measure of the structural changes of the lattice, increases with the Fe concentration (Fig. 4), in agreement with the expansion of the lattice displayed by X-ray diffraction.

For the samples in the range  $0 \leq x \leq 0.3$ , the low temperature Mössbauer spectra are very similar with the spectra at room temperature, which can be interpreted as absence of magnetic fluctuation or magnetic ordering in this range of Fe concentration, even at 4.2 K. These results seem to contradict the expectation of simple band filling, according to which magnetic fluctuations would be responsible for the disappearance of superconductivity [4,5]. On the other hand, for  $x \geq 0.3$  the low temperature Mössbauer spectra broaden and a magnetic hyperfine field is detected at the Fe nucleus

(Ni site) (Fig. 3). Then Mössbauer and ac susceptibility results indicate that a magnetic ordering or magnetic fluctuations is established only for Fe concentrations far away from that for which the superconductivity is completely destroyed ( $x \sim 0.04$ ). Further experiments, of Mössbauer spectroscopy under external field in the sample with 1 at. % of Fe, are in progress to confirm the presence of magnetic moment at the Fe.

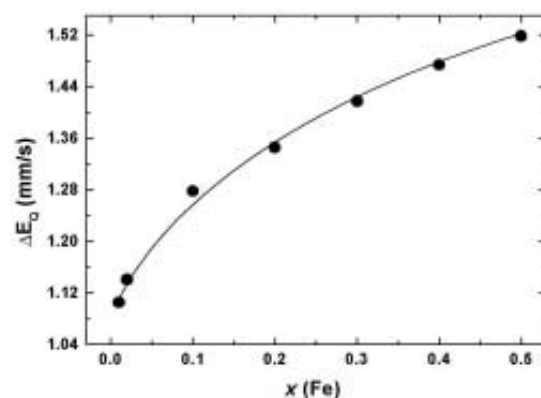


Figure 3. Room temperature electrical quadrupole splitting  $\Delta E_Q$  observed in  $\text{MgC}(\text{Ni}_{1-x}\text{Fe}_x)_3$  ( $0 \leq x \leq 0.3$ ).

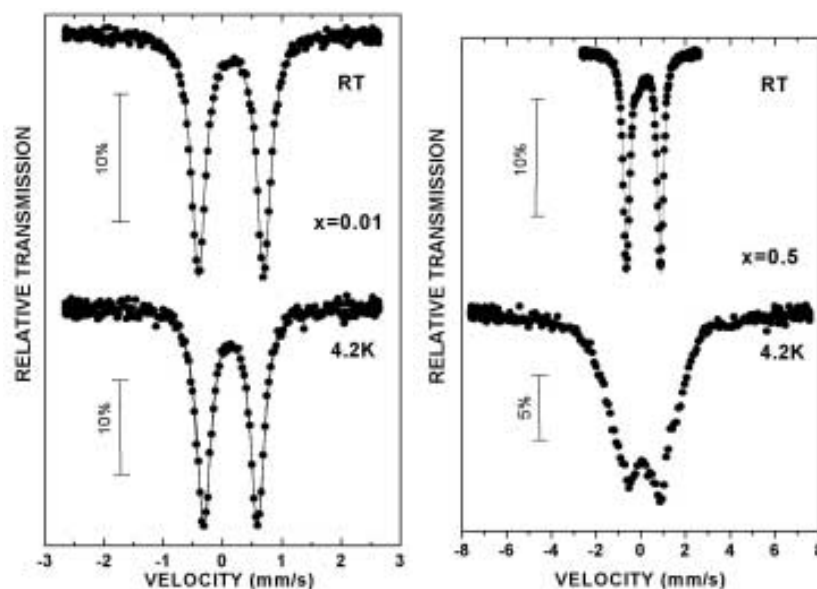


Figure 4. Room temperature and 4.2 K Mössbauer spectra of the samples  $\text{MgC}(\text{Ni}_{1-x}\text{Fe}_x)_3$  with  $x = 0.01$  and  $x = 0.5$ .

Band structure calculations in  $\text{MgC}(\text{Ni}_{1-x}\text{Co}_x)_3$   $x \leq 0.03$  (assuming the same structural parameter as in  $\text{MgCNi}_3$ ) [6] yield no magnetic instabilities, in agreement with the experiments where no evidence of magnetic or structural transition has been reported yet for Co doping [6], showing that the simple band filling

model may not work well in the case of Ni substitution. However, an expansion of the lattice with the Co doping is also expected, similar to that produced by Fe doping (above), and must be considered in the band structure calculation. On the other hand, structural effects on the electron-phonon coupling may play an important role in

describing the nature of the depression of  $T_c$ . Hence, the structural effects, due to the Ni doping, should be included in the analysis of the ground state properties of these series.

## IV Conclusion

Our preliminary results on X-ray diffraction,  $^{57}\text{Fe}$  Mössbauer spectroscopy, on the  $\text{MgC}(\text{Ni}_{1-x}\text{Fe}_x)_3$   $0 \leq x \leq 0.5$  system show that an expansion of the lattice with the Fe doping occurs. This expansion was monitored through the change in the lattice parameter and the electrical quadrupole splitting  $\Delta E_Q$ . Preliminary ac susceptibility results show no evidence of magnetic fluctuation and no magnetic ordering was observed neither in the range of Fe doping for which the superconductivity was systematically destroyed ( $x \leq 0.04$ ) nor just above of that, contrary to the expected by simple band filling model. Only for concentration well above of the value for which the superconductivity is completely destroyed ( $x \geq 0.3$ ), the establishment of a magnetic ordered state is observed by ac susceptibility as by Mössbauer spectroscopy. These results suggest that Fe has a magnetic moment in this system.

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