

Mössbauer Studies on the Intermetallic Pseudo-Ternary
 $Ce(Ru_{1-x}Fe_x)_2Ge_2$ Compound ($0 \leq x \leq 1$)

M. B. Fontes, E. C. Passamani, M. El-Massalami and E. M. Baggio-Saitovitch

Centro Brasileiro de Pesquisas Físicas

Rua Dr. Xavier Sigaud 150

Urca, CEP:22290-180

Rio de Janeiro - Brazil

ABSTRACT

^{57}Fe Mössbauer measurements on the $Ce(Ru_{1-x}Fe_x)_2Ge_2$ series of compounds ($0 \leq x \leq 1.0$) were performed at 300 K and 4.2 K. All samples showed a quadrupolar interaction with an asymmetry that was due to texture effects. Moreover, at 4.2 K, and for $x < 0.5$, the observed spectra could be well fitted with a small transferred hyperfine field (< 1 T) that decreased with Fe -concentration. This agrees with our *ac* susceptibility observation that the localized character of the Ce -moment was systematically decreasing with Fe concentration. For $x > 0.5$, no transferred hyperfine field was observed, consistent with the absence of localized Ce -moments supported by our magnetization measurements.

Key-words: $Ce(Ru_{1-x}Fe_x)_2Ge_2$, Intermetallic Compounds, Mössbauer Spectroscopy, Magnetic Properties,

Introduction:

The family of compounds RM_2X_2 (R = rare earth or actinide, M = transition metal and $X = Si, Ge$) crystallizes in the $ThCr_2Si_2$ structure (space group $I4/mmm$) [1]. It was reported to show a variety of interesting physical properties such as: superconductivity, Kondo effect or heavy fermion behavior. As an example, $CeCu_2Si_2$ is a heavy fermion superconductor ($T_c = 0.6$ K) [2], $CeRu_2Si_2$ is a heavy fermion ($T_K = 20$ K) [3], $CeRu_2Ge_2$ is a ferromagnet ($T_c = 7.5$ K) [4] and $CeFe_2Ge_2$ is a Pauli paramagnet with no localized moment on Ce nor on Fe -ion.

We are currently interested in studying how the Ce magnetism respond to partial substitution of $M = Ru$ by Fe atoms in the parent compound $CeRu_2Ge_2$. XRD structural studies, ac susceptibility, magnetization, resistivity, Mössbauer spectroscopy (ME) were utilized for this investigation. The analysis of the XRD data showed that the Fe ions enter substitutionally in the whole concentration range and within the XRD sensitivity, no spurious phases were detected. In this communication, we report only on the Mössbauer spectroscopy results. For these series, the Mössbauer spectra showed a quadrupole interaction in the whole temperature and concentration range. Moreover, at 4.2 K, a small hyperfine field (presumably transferred from Ce -moments) was observed for $x < 0.5$. For this range ac susceptibility and magnetization measurements showed Curie-Weiss behavior which was due to the localized moment on the Ce -ion.

Experimental:

Samples of $Ce(Ru_{1-x}Fe_x)_2Ge_2$ with $x = 0, 0.005, 0.015, 0.03, 0.05, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9$ and 1.0 were prepared by arc-melting the stoichiometric amount of high purity elements in an argon arc-furnace. Later, they were annealed at $1000^\circ C$ for 7 days in a closed quartz tube with partial Ar-pressure, afterwards they were quenched in liquid nitrogen. Powder X-ray diffraction was carried out at 300 K using $Cu-K\alpha$ radiation. Structural analysis were performed using the conventional Rietveld Structural procedures. Mössbauer spectra were taken at 4.2 K and 300 K, in transmission geometry and constant acceleration mode, with a 25 mCi ^{57}Fe in Rh matrix. Source and absorber were kept at the same temperature during the measurements. The isomer shift (IS) values are relative to α - Fe absorber.

Results and Discussion:

The X-ray analysis showed that all the samples were single phase with the expected tetragonal $ThCr_2Si_2$ ($I4/mmm$) structure. Lattice parameters (a, c) as well as the unit cell volume depend monotonically on the Fe -substitution (a decreases with x while c increases,

but the overall change leads to a decrease in the unit cell volume) and, as expected, scales with the atomic radii of *Ru* (1.34 Å) and *Fe* (1.26 Å).

Mössbauer spectra, with unresolved quadrupole doublets are shown in fig. 1.a (T=300 K) and fig. 1.b (T=4.2 K). Each of the spectra was well fitted with symmetrical doublet of lorentzian line shapes; the line widths were found to be 0.26(2) mm/s while the *IS* and *QS* are shown in fig. 2.a and fig. 2.b. The Mössbauer spectra at 4.2 K are more complex because they are the combination of quadrupole and magnetic interactions of similar magnitude, and therefore an analysis was performed using a complete hamiltonian for diagonalization. The line widths obtained from the fits were around 0.28(2) mm/s, while the *IS*, *QS* and B_{hf} values are plotted in fig. 2.a, fig. 2.b and fig. 2.c. The variation of the *IS* with composition is the same for 300 K and 4.2 K measurements and shows a clear decrease of *IS* with *Fe* content. The observation of a magnetic hyperfine field (B_{hf}) for $x \leq 0.5$ is consistent with localized magnetism of the *Ce*-moments as observed by *ac* susceptibility measurements. The apparent discrepancy of *QS* values at 300 K and 4.2 K for $x \leq 0.5$ is due to the angular factor which was not considered in the 4.2 K analysis. In fact there is a change of the angle θ between B_{hf} and *EFG* with the composition. If we fix the *QS* to the *RT* values, θ will vary in the range of $31^\circ \leq \theta \leq 55^\circ$.

The overall concentration dependence of Mössbauer parameters can be summarized as follows: The line widths are sharp (0.26-0.28 mm/s) suggesting good sample homogeneity. The quadrupole splitting (and hence the *EFG* at the *Fe*-nucleus), increases with concentration up to $x = 0.5$ beyond that it remains constant at 0.2 mm/s. The *IS* decreases monotonically with concentration showing that the *S*-electronic density at the *Fe* nucleus is increasing with x . For $x < 0.2$, the B_{hf} (T= 4.2 K) was found to be 1.3 T, decreasing sharply to zero within our limit of experimental discrimination for $x > 0.5$. It is worth emphasizing that for all concentrations, a single Mössbauer *Fe*-site was found indicating that the *Ru* is substituted by *Fe* in equivalent positions. In the following, we will try to interpret the concentration dependence of the *IS*, *QS* and B_{hf} .

From crystal structure, the *Fe* ions are reported to reside in *4d* site and are tetrahedrally coordinated to four *Ge* ions (*4e* site) as nearest neighbors, and to four *Ce* ions (*2a* site) as next nearest neighbors [1]. Any deviation from this regular coordination will generate a small lattice contribution to the quadrupolar interaction at the *Fe* site [5]. Rietveld structural analysis shows that *Fe*-substitution leads to a compression along the basal plane and an elongation along the *c*-axis, consequently leading to a rearrangement of bonds lengths and bond angles of all ions in the unit cell. It is highly possible that the observed *QS* concentration behavior is related to this lattice modification. As well known, pure ionic rearrangement may strongly influence the *EFG* at the *Fe*-site and thus the *QS*. On

the other hand, the *IS*-concentration dependence may be attributed to an increase in the density of electrons at the *Fe*-nucleus due to a decrease in the unit cell volume with *Fe*-substitution.

Magnetization and *ac* susceptibility studies on this series showed that the localized character of the *Ce* 4*f*-moments systematically decreases with *Fe*-concentration: T_C decreases monotonically for $0 \leq x \leq 0.7$. Above $x = 0.7$, only Pauli paramagnetism was observed. Based on these results and also in the fact that the values of B_{hf} are small indicates that *Fe* has no moment in this compound and B_{hf} is transferred from *Ce*-moments. This assumption is strongly supported by the fact that there is no correlation between *IS* and B_{hf} .

Conclusions:

^{57}Fe Mössbauer spectroscopy analysis of $\text{Ce}(\text{Ru}_{1-x}\text{Fe}_x)_2\text{Ge}_2$ series of compounds is consistent with single site for *Fe* substituting *Ru* ions. Both *QS* and *IS* concentration variation is due to a modification in the electronic structure of the *Fe*-ions induced by the unit cell volume contraction with *Fe* content. *Fe* has no moment in this series and a small transferred hyperfine field was observed for samples, that displayed magnetic ordering of *Ce*-moments ($x \leq 0.5$); for higher *Fe*-concentration, *Ce*-moment was found to decrease sharply and moreover no B_{hf} is detected at *Fe*-site.

Acknowledgements:

We appreciate the experimental help of R. Pereira. The work was supported by *RHAE/* New Materials Program (*MCT*) and *CNPq* grant #500938/91-1.

Figure Captions:

Fig.1. ^{57}Fe Mössbauer spectra for $\text{Ce}(\text{Ru}_{1-x}\text{Fe}_x)_2\text{Ge}_2$ at (a) 300 K and (b) 4.2 K.

Fig.2 (a) B_{hf} - *x*, (b) *IS* - *x* and (c) *QS* - *x* for $\text{Ce}(\text{Ru}_{1-x}\text{Fe}_x)_2\text{Ge}_2$ obtained at 4.2 K (⊙) and 300 K (Δ).

Fig.1

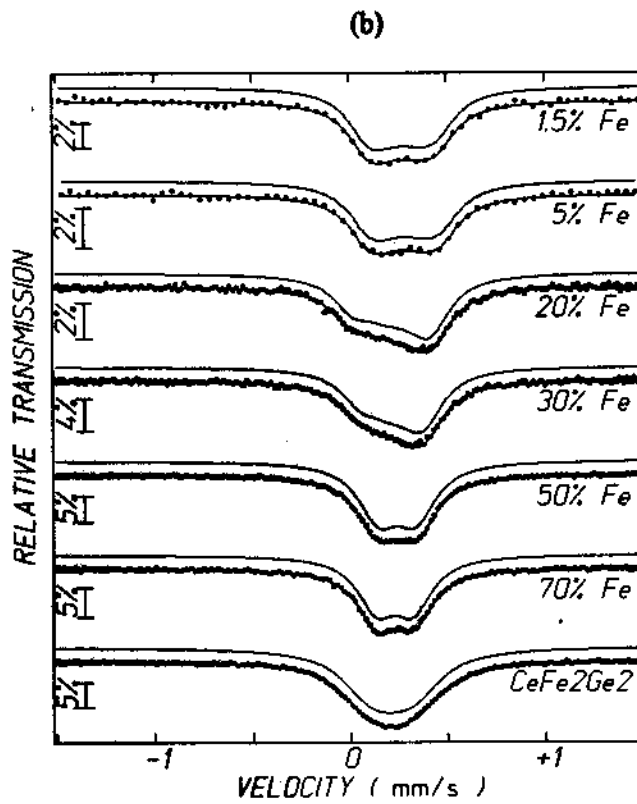
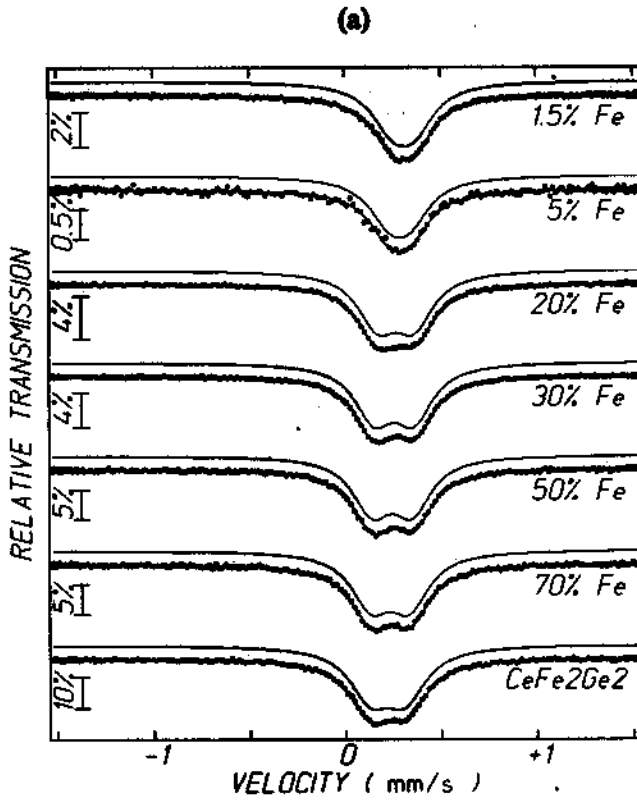
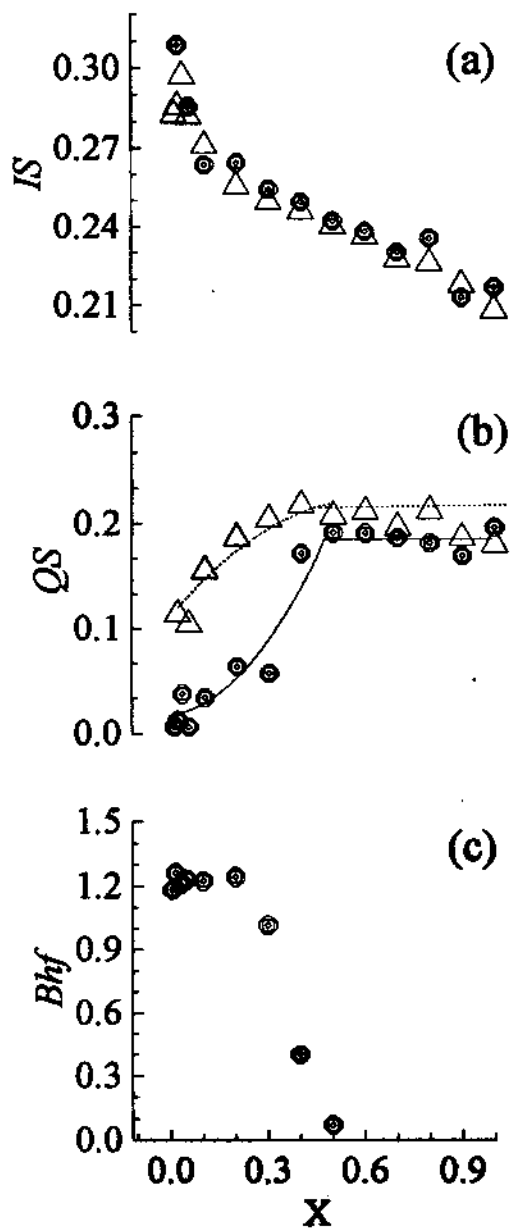


Fig.2



References:

- [1] Z. Ban and M. Sikirica, *Acta Cryst.* **18** (1965) 594
- [2] F. Steglish, J. Aarts, C. D. Bredl, W. Lieke, D. Meschede, W. Franz and H. Schäfer, *Phys. Rev. Lett.* **25** (1979) 1892.
- [3] L. C. Gupta, D. E. Mac Laughlin, Cheng Tien, C. Godart, M. A. Edwards and R. D. Parks, *Phys Rev. B.* **28** (1983) 3673
- [4] M. J. Besnus, A. Essaihi, N. Hamdaoui, G. Fisher, J. P. Kappler, A. Meyer, J. Pierre, P. Haen and P. Lejay, *Physica B* **171** (1991) 350.
- [5] A. M Umarji, D. R. Noakes, P. J. Viccaro, G. K. Shenoy, A. T. Aldred and D. Niarchos, *J. Magn. Magn. Mat.* **36** (1983) 61-65