Low-Temperature Properties of $Ce(Ru_{1-x}M_x)_2Ge_2$, M = Fe, Au

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ABSTRACT

We have performed measurements of electrical resistivity, AC susceptibility, magnetization, Mössbauer effect and NMR on the series of intermetallic compounds $Ce(Ru_{1-x}M_x)_2Ge_2$, M = Fe, Au for $0 \leq x \leq 0.1$. The parent compound $CeRu_2Ge_2$ orders ferromagnetically below 7.5 K, with f-electrons showing only a small mass-enhancement at low temperatures. However, its crystal structure and Fermi surface are closely related to the heavy-electron superconductor $CeRu_2Si_2$. Analysis of X-ray diffraction patterns confirmed the structure to be of the $ThCr_2Si_2$-type, with no foreign phases detected. The results of our Mössbauer studies indicate that Fe goes into the Ru sites with no magnetic moment. NMR signals were observed in the Fe-doped samples with quadrupolar interaction that can be related to ^{73}Ge nucleus (I = 9/2).

Key-words: $Ce(Ru_{1-x}M_x)_2Ge_2$, magnetic properties, resistivity, NMR.

1. Introduction

The series CeM_2X_2 (M: transition metal, X: Ge or Si) with the tetragonal $ThCr_2Si_2$-type structure [1] has a variety of interesting physical properties: $CeCu_2Si_2$ is a superconducting heavy fermion system [2] while $CeRu_2Si_2$ is a heavy fermion system with non-magnetic ground state [3]. On the other hand, $CeRu_2Ge_2$ [4] is a ferromagnet, $CeFe_2Ge_2$ [5] is a Pauli paramagnet, and $CeAu_2Ge_2$ is a normal antiferromagnet below 15 K [6]. $CeRu_2Ge_2$ is interesting because although the Fermi surfaces of both $CeRu_2Si_2$ and $CeRu_2Ge_2$ are very similar, the electronic mass enhancement in $CeRu_2Ge_2$ is just three times that of free electrons [7]. Moreover, it orders ferromagnetically below . T_C = 7.5 K and its Kondo temperature is estimated to be lower than 1 K [6]. Above T_C the localized effective moment of Ce is around 2 μ_B [7,8]. In its polycrystalline form, a second transition around 8.5 K can be observed, the origin of which is not well known up to now [4,6].

In this work, we study the changes in the magnetic and transport properties of $CeRu_2Ge_2$ induced by the substitution of a magnetic (Fe) or non magnetic (Au) atoms on the Ru-site. The transport and magnetic properties of $CeRu_2Ge_2$ do not change significantly with the addition of small amount of impurities of Fe or Au. The formation of softer domain walls in the Fe-doped samples is argued to be the mechanism which allows the observation of NMR signals from ^{73}Ge.

2. Experimental

Samples were prepared by melting stoichiometric amounts of high purity elements in an argon arc-furnace, annealed for 7 days at 1000°C in 1/3 argon atmosphere in a closed quartz tube and later were quenched in liquid nitrogen. Eleven different samples were prepared: $Ce(Ru_{1-x}M_x)_2Ge_2$ with M = Au, Fe and x = 0, 0.005, 0.015, 0.03, 0.05 and 0.1. X-ray data were taken at room temperature using Cu Kα-radiation. Lattice parameters were obtained using the conventional Rietveld Structural Analysis program.

DC resistivity was measured using the standard four points technique. Magnetic properties were measured by DC magnetization and AC susceptibility. NMR was measured at 4.2 K using an automated pulsed spectrometer. ^{57}Fe Mössbauer spectra were obtained at room temperature and 4.2 K using transmission geometry, with a ^{57} Co-Rh source kept at the same temperature.

3. Results and discussions

All the studied samples doped with Fe show single phase with the expected tetragonal $ThCr_2Si_2$ structure and with a systematic concentration dependent change in the lattice parameters. This indicates that Fe enter the structure substitutionally. Additional information obtained by Mössbauer spectroscopy indicates that Fe occupies the Ru site and has no

magnetic moment, however a small transferred hyperfine field (≈ 1 T) was found. In the case of gold substitution the situation is more complex, since for samples with x=0.1 spurious phases were observed in the X-ray measurements.

The temperature dependence of ρ, for all samples, is very similar to ρ(T) of $CeRu_2Ge_2$ [4]. However, the temperature-independent resistivity increases with the addition of impurities (Fe or Au) as expected from Matthiessen's rule. The Curie temperature T_C (≈7.5 K) and the transition width (ΔT≈4K) are independent of the Fe and Au concentration, which shows that the magnetism of the host is not affected by the Ru substitution. However, the influence on the dynamics of the domain walls due to the Fe in the matrix is clearly seen in the results of AC susceptibility measurements: the drop in χ_{ac}(T) below T_C, and the observed frequency dependency of χ'' are characteristic of soft domain walls motion (Fig. 1). Finally, the zero-field cooling (ZFC) and field-cooling (FC) magnetization of the 5% of Fe show a splitting of the M vs. T curve below T_C (Fig. 2). This splitting is characteristic of magnetic samples with soft domain walls.

NMR signal was observed, so far, only for the Fe-doped samples. The central frequency is roughly independent of the Fe-concentration (up to 5% of Fe). The signal intensities are very weak, and are observable only after suitable averaging of many spin-echo events. However, samples with higher Fe concentration show larger NMR signals with a well-resolved structure (Fig. 3). This spectra correspond to a first order perturbation of the Zeeman energy-levels by the presence of a small electric quadrupolar interaction for a spin 9/2, as is the case for ^{73}Ge nuclei in $Ce(Ru_{1-x}\,Fe_x)_2Ge_2$. Assuming that the quadrupolar interaction has axial symmetry, as expected from crystal structure, we estimate the hyperfine field at the Ge sites ($H_{hf} \approx 440$ kG). The main contribution arises from the ordered magnetic moments of the Ce 4*f*-electrons. The large amplification factor needed to observe the NMR signal from the ^{73}Ge nuclei is provided by the soft domain walls of $Ce(Ru_{1-x}\,Fe_x)_2Ge_2$. This was confirmed by observing that the NMR signals disappear at low-applied magnetic fields.

4. Conclusions

The results of our measurements indicate that the electronic structure of $CeRu_2Ge_2$ is very stable with respect to the substitution of Ru by small amounts of Fe or Au. The Ce moments are well localized and they are far from moment instabilities. This only adds to the puzzle about $CeRu_2Ge_2$ and $CeRu_2Si_2$; while having similar crystalline and electronic structures, their Ce 4*f*-electrons behave very differently at low temperatures.

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Figure Captions:

Fig.1 - AC susceptibility for $Ce(Ru_{0.95}Fe_{0.05})Ge_2$ showing the real part χ' and the imaginary part χ''.

Fig. 2 - ZFC and FC magnetization measurement for $Ce(Ru_{0.95}Fe_{0.05})Ge_2$.

Fig. 3 - NMR spectra for $Ce(Ru_{0.95}Fe_{0.05})Ge_2$ at 4.2 K. The spectra are normalized and the continuous line connects the experimental data points.

Fig.1

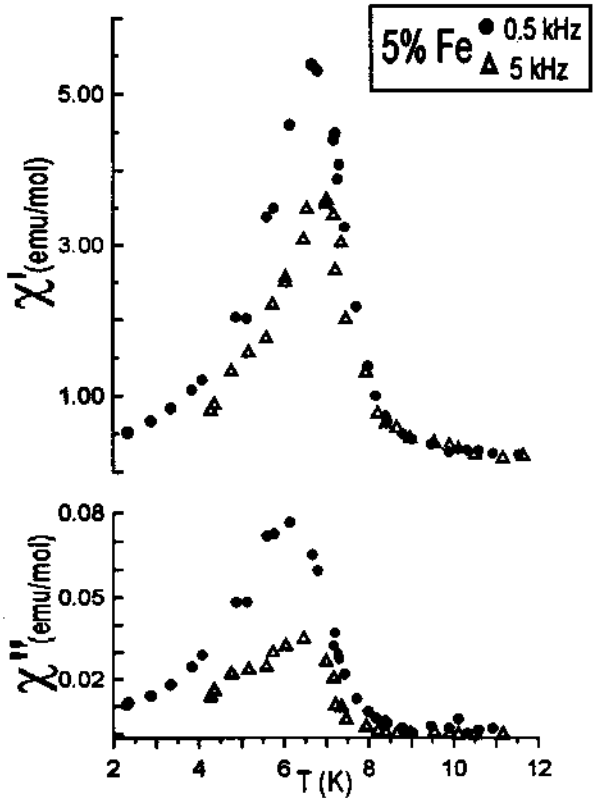

5% Fe
0.5 kHz
5 kHz
χ'(emu/mol)
χ''(emu/mol)
T (K)

Fig.2

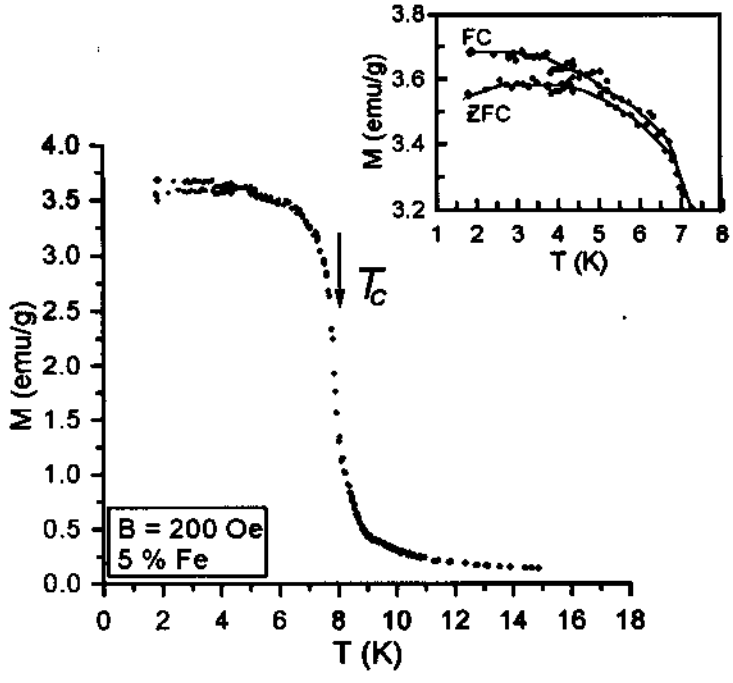
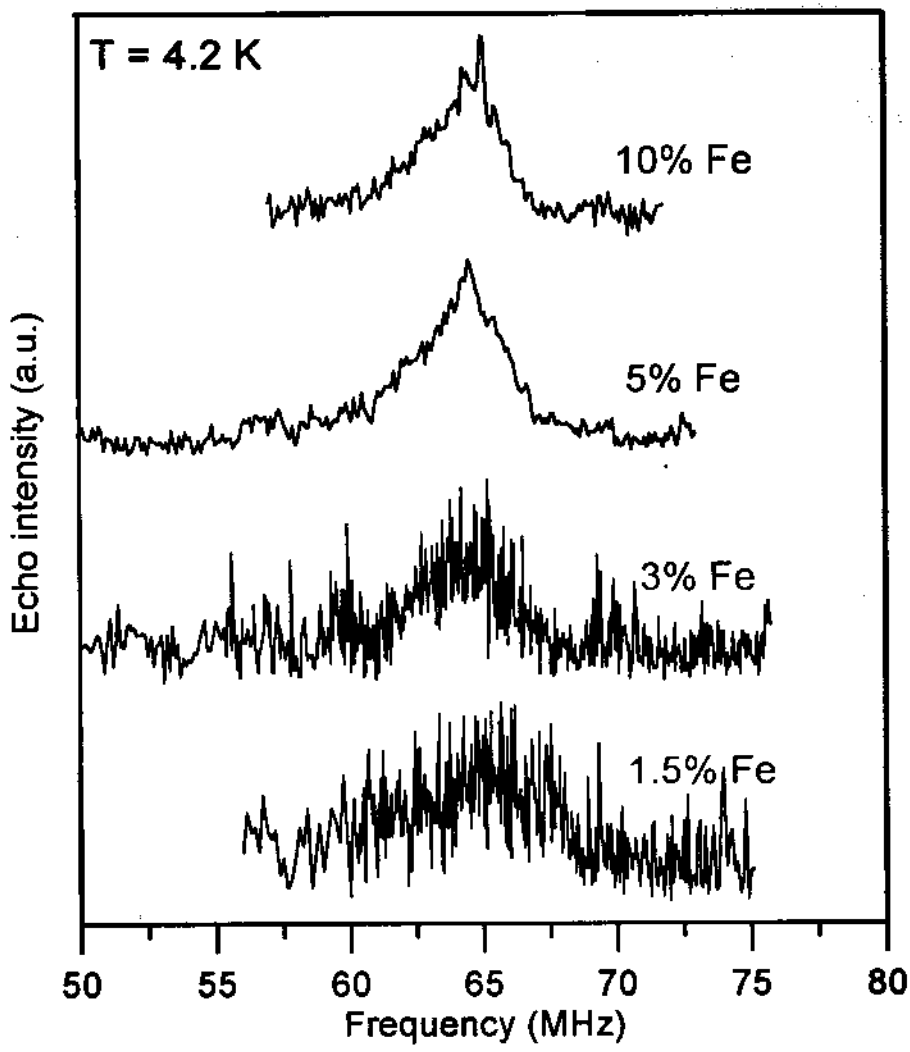

FC
ZFC
M (emu/g)
T (K)
T_c
B = 200 Oe
5 % Fe

Fig.3


T = 4.2 K
10% Fe
5% Fe
3% Fe
1.5% Fe
Echo intensity (a.u.)
50
55
60
65
70
75
80
Frequency (MHz)

References:

[1] Z. Ban and M. Sikirica, Acta Cryst. **18** (1965) 594

[2] F. Steglich, J. Aarts, C. D. Bredl, W. Lieke, D. Meschede, W. Franz and H. Schäfer, Phys. Rev. Lett. **43** (1979) 1982

[3] L. C. Gupta, D. E. Mac Laughlin, Chem Tien, C. Godart, M. A. Edwards and R. D. Parks, Phys. Rev. B **28** (1983) 3673

[4] M. J. Benus, A. Essaihi, N. Hamdaoui, G. Fisher, J. P. Kappler, A. Meyer, J. Pierre, P. Haen and P. Lejay, Physica B **171** (1991) 350

[5] J. J. Bara, H. U. Hrynkiewicz, A. Milos and A. Szytula, J. Less-Common Met. **161** (1990) 185

[6] A. Loidl, G. Knopp, H. Spille, F. Steglich and A. P. Murani, Physica B **156&157** (1989) 794

[7] C. A. King and G. G. Lonzarich, Physica B **171** (1991) 161

[8] A. Böhm, R. Caspary, U. Habel, L. Pawlak, A. Zuber, F. Steglich and A. Loidl, J. Magn. Magn. Mat. **76&77** (1988) 150