ESRF	**Experiment title:** A ^{57}Fe and ^{151}Eu nuclear resonance scattering study of RFeAs(O,F) and $EuFe_2As_2$	**Experiment number**: Group of HE 3049 3050 and 3052
Beamline: ID18	**Date of experiment**: from: March 4th 2009 to: March 18th 2009	**Date of report**: Sept 4th 2009
Shifts: 36	**Local contact(s)**: I. Sergeev	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

R. P. Hermann,[1,2,*] M. Angst,[1,*] W. Schweika,[1,*] R. Mittal,[1,*] U. Pelzer,[3,*] I. Sergeev,[3,*] M. A. McGuire[4]
1. Institut für Festkörperforschung, JCNS und JARA-FIT, Forschungszentrum Jülich, Jülich, Germany
2. Departement de Physique, Université de Liège, Liège, Belgique
3. European Synchrotron Radiation Facility, Grenoble, France
4. Condensed Matter Division, Oak Ridge National Laboratory, Oak Ridge TN, USA

Report:

We have carried out measurements of the nuclear inelastic scattering, respectively by iron-57 and europium-151 in $RFeAsO_{1-x}F_x$ (R = La and Nd) and $EuFe_2As_2$ in order to obtain the partial phonon density of states. The measurements were carried out at several relevant temperatures and pressures, notably, above and below the superconducting phase transition in the F doped compounds. Further, we have measured the nuclear forward scattering by iron-57 and europium-151 at selected temperatures and pressures.

These measurements were carried in the context of the recent discovery of superconductivity, at a temperature T_c higher than all non-copper oxide superconductors in iron based compounds, most notably, in fluorine-doped *R*FeAsO (*R* = rare earth) and K-doped $BaFe_2As_2$, a discovery which has attracted extensive attention in the scientific community since April 2008 [1,2]. The mechanism of superconductivity, and in particular the role of lattice dynamics, is still unclear. Superconductivity in these compounds is believed to be mainly due to the structural and electronic states of the Fe-As layers, in relation with magnetic fluctuations. It has been suggested that the in plane Fe phonon breathing modes are important for the superconductity[3] however it was rapidly shown that these Fe-As based compounds do not obey the classical electron-phonon paradigm for superconductivity[4]. Since our measurements, in March 2009, there was however several reports about the observed isotope effect on the superconducting transition temperature[5], that clearly indicate that next to magnetic excitations[6], the lattice dynamics plays an important role.

The first series of experiments was dedicated to the $RFeAsO_{1-x}F_x$ compounds, with R = La and Nd, and $x = 0$ or 0.1. All samples were 95% iron-57 enriched. Measurements of the iron phonon DOS revealed that the general behaviour is a simple scaling of all phonon energies, and after the proper linear scaling of the energy spectra at all temperatures and pressures can be superimposed; further, the phonon DOS in the F doped and parent compounds are extremely similar. However, very interestingly, two anomalous behaviours were observed. First, one particular peak in the iron partial DOS, at ~16 meV, exhibits a significant shift upon doping with F. Second, this particular phonon peak also exhibit a shift as a function of temperature, with

respect to the other phonon modes. It must be noted that measurements similar to those reported herein were carried out earlier at SPRing-8[7] with however a worse energy resolution (ca. 2 meV) which prevented to observe the significant shifts in phonon energies that we observed here, with an energy resolution of 0.7 meV. We could not detect any particular modification in the phonon DOS of the F doped compounds upon undergoing the superconducting transition. Likely such effect is small, and measurements with a much better resolution, around 0.1 meV would be necessary to resolve these effects. Likewise, our careful investigation of the nuclear forward scattering around the superconducting temperature didn't either reveal any modification in the hyperfine interactions.

The second and third series of experiments were dedicated to the study of the iron and europium magnetism and lattice dynamics in $EuFe_2As_2$. The particular motivation for this study is related to the possible pressure induced reentrant superconductivity in undoped $EuFe_2As_2$[8]. One sample was 95% iron-57 enriched and polycrystalline, whereas a second sample was a natural abundance single crystal. Our nuclear inelastic scattering measurements revealed a similar simple scaling behavior as in $RFeAsO_{1-x}F_x$, with however a significant anisotropy of one particular iron phonon mode, at ~15 meV, as revealed by scattering on a single crystal with the incident beam in- and out-of- the *a-b* plane. Further interesting results are, first, the absence of coupling between the rare-earth and the iron magnetic sublattices: the nuclear forward scattering by iron-57 around the Eu ordering temperature indicates no modification in the iron hyperfine field, see Figure 1. Second, despite a detailed investigation of the pressure/temperature range in which reentrant superconductivity was proposed (P>2.5 Gpa and T~25-30 K) we could not find the signature of such a transition: we observe the expected gradual suppression of the iron magnetism as a function of pressure, evidenced by the gradual decrease of the spin-density-wave transition, but we did not observe any modification of the magnetically split hyperfine iron spectrum upon entering the supposed superconducting phase. We conclude that either no real bulk superconductivity occurs, or, that the iron hyperfine field is not expelled in such phase.

In summary, high-resolution nuclear inelastic scattering appears to be a highly important technique for resolving subtle phonon behaviour in the Fe based superconductors, in particular because in contrast to the equally well resolved Raman spectroscopy, it is not restricted to the Γ point, and because in contrast to inelastic neutron scattering, that suffers from overlapping Fe and As phonon modes, the element specificity is warranted. In this light, improving the resolution of the nuclear inelastic monochromators for iron-57 appears to be vital.

References

[1] Y. Kamihara *et al.*, J. Am. Chem. Soc. **130**, 3296 (2008).
[2] Ishida K. *et al.*, J. Phys. Soc. Jap. 78, 062001 (2009).
[3] Eschrig H., http://arxiv.org/abs/0804.0186
[4] Boeri L. *et al.*, Phys. Rev. Lett. **101**, 026403 (2008).
[5] Liu R. H. *et al.*, Nature **459**, 64-67 (2009).
[6] Christianson A. *et al.*, Nature **456**, 930-932 (2008).
[7] Higashitaniguchi S. *et al.*, Phys. Rev. B **78**, 174507 (2008).
[8] Miclea C. F. *et al.*, Phys. Rev. B **79**, 212509 (2009).