Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office using the **Electronic Report Submission Application:**

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The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

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Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
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Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** **Eu-151 NFS high-pressure study of magnetism in CsCl-type EuS and EuSe**	**Experiment number**: HE-1490
Beamline: ID22N	**Date of experiment**: from: 07.05.03 to: 13.05.03	**Date of report**: 30.08.03
Shifts: 18	**Local contact(s)**: **Dr. Alessandro Barla**	*Received at ESRF:*

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Report: This experiment is a continuation of our proposal HE-592, where we performed in April 1999 the first nuclear-forward scattering (NFS) experiments with the 21.5 keV Mössbauer radiation of Eu-151 on EuTe and EuS in the CsCl-type high-pressure phase [1, 2].

In the present experiment we extended our investigations on the magnetic behaviour of the Eu(II)-chalcogenides in their CsCl-phases to EuSe and studied EuS at much higher pressures. We used now advanced focusing optics (compound refractive lenses and a bending crystal) to concentrate the whole monochromatized synchrotron radiation on the small samples contained in a diamond anvil cell (DAC). The DAC was mounted in a He cryostat with the option to apply an external magnetic field to the sample. By an external field the NFS spectra become more simple and one obtains, in addition, information on the ferromagnetic (fm) or antiferromagnetic (afm) ordering type of the investigated sample [1].

We measured NFS spectra at 300 K with EuF_3 and/or EuS as reference absorbers to obtain isomer shifts S_{IS} [3] and at lower temperatures to obtain magnetic information from the observed magnetic hyperfine fields B_{hf} (T) of EuS up to 77 GPa and of EuSe up to 48 GPa (see Fig. 1). The ferromagnetic ordering temperatures derived from these spectra are T_C = 183(1) K for EuSe and T_C = 290(5) K for EuS. The occurrence of ferromagnetic order was proved in additional NFS studies with an external field, similar to our previous studies of CsCl-type EuTe [1,2]. The simple beat patterns at low temperatures (Fig. 1, 3 K) indicate a strong pressure-induced magnetic texture in the samples.

In Fig. 2 we plotted the derived values of S_{IS}, B_{hf} ($T \rightarrow 0$ K) and T_C in dependence of the applied pressure for EuS. We attribute the strong increase of the isomer shift above 50 GPa to a transition to a strongly mixed-valent state of the Eu ions, in accordance with previous reflectivity measurements reporting the onset of mixed-valent behaviour in EuS around 35 GPa [4]. Such a mixed valence was actually expected in our proposal and we performed therefore the isomer shift measurements. The (preliminary) valence scale v(S_{IS}) in Fig. 2 was obtained by a linear scaling of the isomer shifts between EuS and EuF_3 at ambient pressure. In CsCl-phase EuTe, EuSe and EuS we observe at lower pressures anomalous small and slightly decreasing values of B_{hf}, but for EuS above 50 GPa a strong increase of B_{hf}, which points also to a mixed-valent state with significant contributions to B_{hf} from the trivalent Eu state above 50 GPa. Interestingly the values of S_{IS}, B_{hf} and T_C in Fig. 2 tend to saturate at the highest pressure reached in this study. A full valence transition to pure trivalent Eu, expected at higher pressures, should result in a dramatic loss of magnetism.

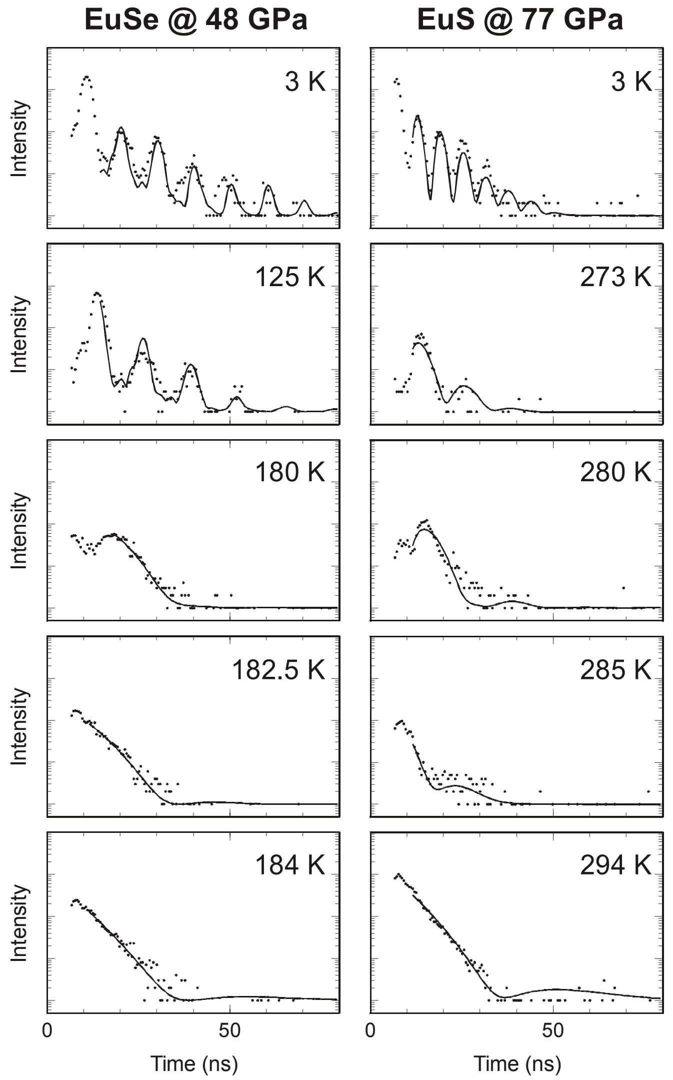

Fig. 1: Eu-151 NFS spectra of CsCl-phase EuSe at 48 GPa and EuS at 77 GPa and various temperatures. The ferromagnetic ordering temperature are determined to 183(1) K for EuSe and to 290(5) K for EuS. The NFS spectra shown at the highest temperatures exhibit only a thickness-induced Bessel minimum, but no magnetic beat structures [1, 2].

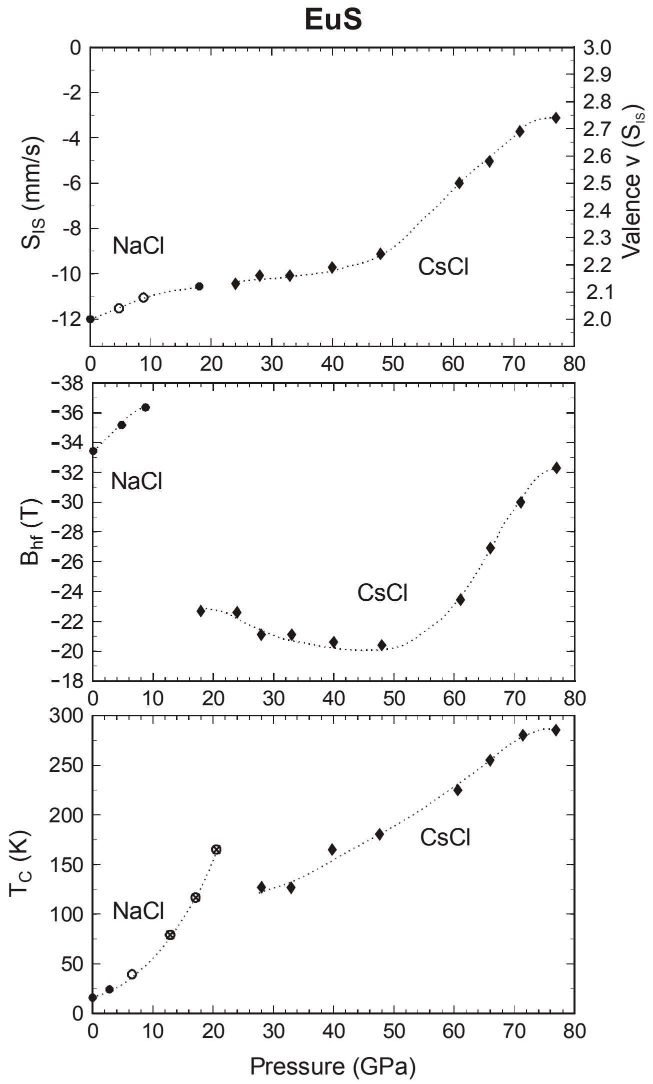

Fig. 2: Isomer shift S_{IS}, magnetic hyperfine field B_{hf} extrapolated to T = 0 K ,and Curie temperature T_C as function of the applied pressure for EuS. The dashed lines are guides to the eye. Data are deduced from: Mössbauer effect (○) [5], neutron diffraction (⊗) [6] and present NFS study (●◆).

In 18 shifts we measured S_{IS}, B_{hf} and T_C for both EuSe and EuS at 5 different pressures (all together about 130 NFS spectra). The evaluation of the experimental data is still in progress; a final interpretation of the data needs also the determination of the lattice parameters of EuS at such high pressures and comparison with divalent (SrS) and trivalent (GdS) reference systems. For this purpose, we are presently performing energy-dispersive XRD studies at HASYLAB in continuation of previous studies of EuS, EuSe and EuTe up to 40 GPa [7]. The outstanding success of this beamtime is based on the improved experimental conditions at beamline ID22N, in particular the focusing optics and the stable experimental conditions provided by the new experimental hutch as well as the longstanding expertise with the Eu-151 resonance at ESRF [1,8].

References: [1] O. Leupold, K. Rupprecht, G. Wortmann, Structural Chemistry 14, 97-107 (2003). [2] R. Lübbers, K. Rupprecht, G. Wortmann, Hyperfine Interactions 128, 115–135 (2000). [3] M. Pleines et al., Hyperfine Interactions 120/121, 181-185 (1999). [4] K. Syassen, Physica 139&140, 277 (1986). [5] J. Moser, G. Wortmann, N. Bykovetz, G.M. Kalvius, J. Mag. Mag. Mat 12, 77 (1979). [6] I.N. Goncharenko and I. Mirebeau, Phys. Rev. Lett. 80, 1082 (1998). [7] K. Rupprecht and G. Wortmann, HASYLAB Report 1999, p.525. [8] O. Leupold et al., Europhys. Lett. 35, 671 (1996).