

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:**

<http://193.49.43.2:8080/smis/servlet/UserUtils?start>

Reports supporting requests for additional beam time

Reports can now be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

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

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

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;
- 1st September for experiments carried out up until January of the same year.

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.



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|---------------------------|----------------------------------------------------------------------------------------------------------|--------------------------------------|
| | Experiment title: Magnetism and valence of EuO in the NaCl- and CsCl-phase up to 92 GPa | Experiment number: HE-2043 |
| Beamline: ID22N | Date of experiment: from: 27.09.05 to: 05.10.05 | Date of report: 28.02.06 |
| Shifts: 18 | Local contact(s): Dr. Ulrich Ponkratz | <i>Received at ESRF:</i> |

Names and affiliations of applicants (* indicates experimentalists):

G. Wortmann*, **K. Rupprecht***, **U. Ponkratz***, **O. Leupold**

Department Physik, Universität Paderborn, D-33095 Paderborn, Germany

U. Ponkratz: ESRF, Grenoble, **O. Leupold:** HASYLAB, Hamburg

Report: The title of this proposal was: "High-pressure phonon spectroscopy of Eu-metal in the bcc and hcp phase". Accordingly we had prepared under inert conditions two Eu-metal samples (from two well-recommended suppliers, stating Eu purity of 99.9% and 99.95%). It turned out, however, that both samples were delivered already partly oxidised. Knowing the isomer shifts and f-factors of Eu-metal, EuO and Eu₂O₃, we could determine in test experiments employing the nuclear forward scattering (NFS) from the beat frequencies of the ¹⁵¹Eu-NFS spectra (Fig. 1a) that the one sample (Chempur) was contaminated with about 10% EuO, while the other sample (Goodfellow) was heavily contaminated with about 20% EuO and Eu₂O₃. With the better sample we did one nuclear inelastic scattering (NIS) scan at 300 K to test the counting rate for the inelastic delayed photons (see Fig. 1b). In the wings of the ¹⁵¹Eu-NIS-spectrum around 10 meV, we had an inelastic count rate of $\approx 5 \text{ s}^{-1}$, which would allow for high-pressure experiments in our high-pressure cells with Be gaskets. High-pressure phonon spectroscopy with a contaminated Eu-metal sample were, however, not performed, because we considered this as a waste of the beamtime. In accordance with the operator and beamline responsible, we therefore switched, after 9 shifts (6 for installing the ¹⁵¹Eu monochromator and the focusing optics, 3 shifts for Eu-metal) to a high-pressure NFS study of EuO up to 92 GPa, in continuation of our successful proposals HE-1490 and HE-1690, where we study the Eu(II)-chalcogenides EuS and EuSe in their CsCl-type high-pressure phases up to the Mbar range [2].

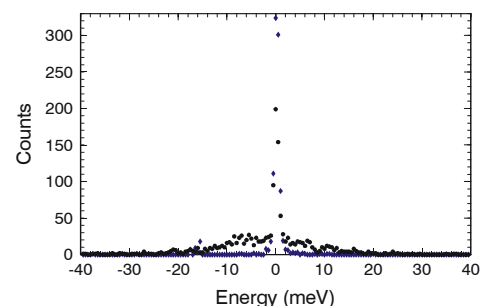
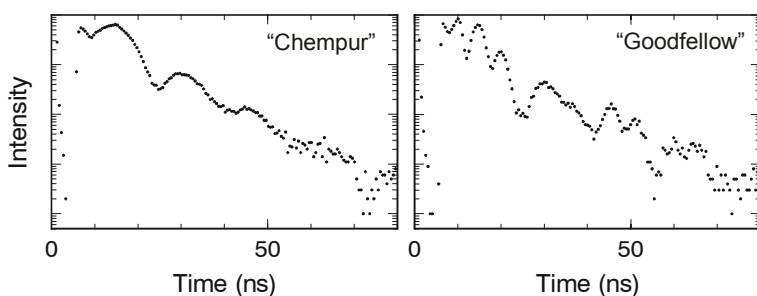


Fig. 1a: Time spectra of Eu-metal with Eu²⁺ and Eu³⁺ impurities from different manufacturers.

Fig. 1b: NIS spectrum of the "Goodfellow" sample at 300 K.

Similar as in our previous ¹⁵¹Eu-NFS studies [1, report HE-1690] we used the advanced focusing optics (compound refractive lenses and bending crystal) to concentrate the whole monochromatized synchrotron radiation on the small samples (80 μm diameter) contained in a miniature diamond anvil cell (DAC). Two DACs loaded with EuO at different pressures were mounted in a new He cryostat equipped with a superconducting magnet in order to apply an

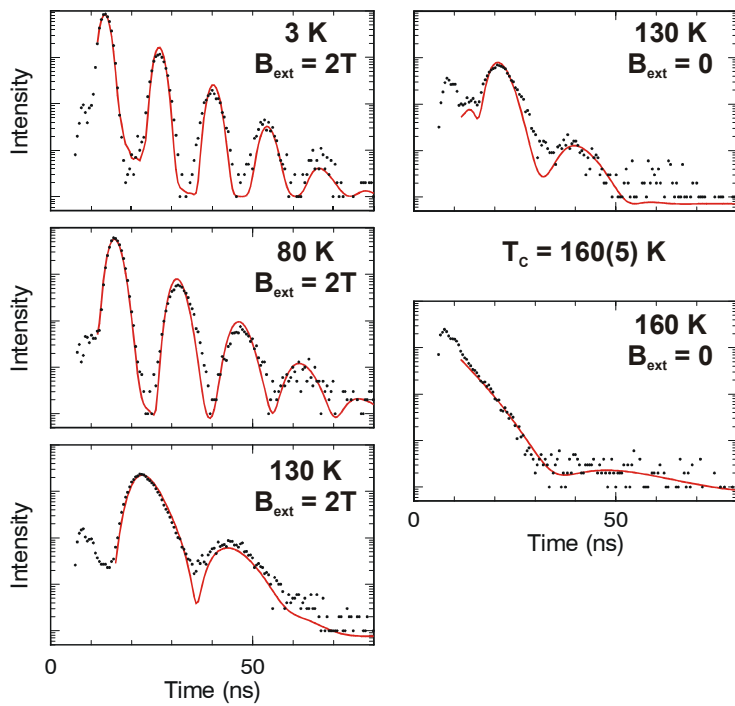


Fig. 2a: Selected ^{151}Eu -NFS spectra of EuO at 69 GPa measured in an external magnetic field of 2T applied perpendicular to the SR direction up to 130 K. Above 130 K the spectra were measured without B_{ext} to determine the magnetic ordering temperature.

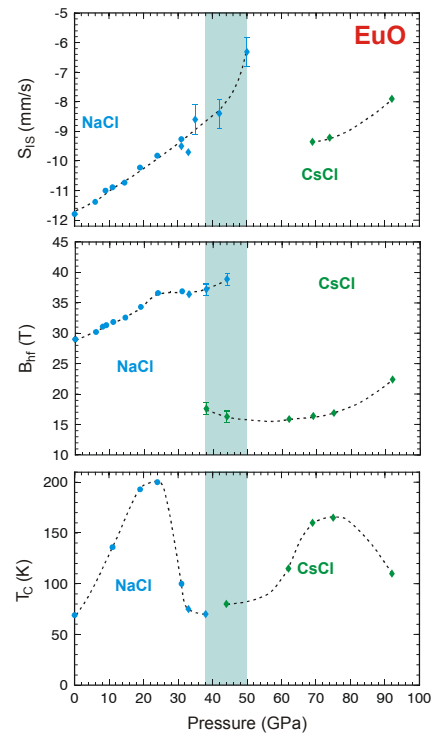


Fig. 2b: The observed parameters S_{IS} , B_{hf} and T_{C} as function of the applied pressure in NaCl- and CsCl-type EuO together with data from [3].

external magnetic field perpendicular to the direction and the polarization of the synchrotron beam. By use of the external field B_{ext} one obtains much simpler NFS spectra and one can determine the magnetic ordering type [1]; we found for EuO, as previously for EuS, EuSe [2] and EuTe [1], ferromagnetic ordering in the CsCl-phase.

The ^{151}Eu -NFS-spectra were recorded in the following sequence: For each new pressure, we measured first at 300 K a NFS spectrum with EuF_3 as trivalent reference absorber in order to obtain the isomer shift S_{IS} of the pressurized sample [1]. Then we went to low temperature (3 K) to obtain the saturation value of the magnetic hyperfine field B_{hf} ($T \rightarrow 0$ K). By measuring NFS-spectra at increasingly higher temperatures, the magnetic ordering temperature T_{C} was determined. Fig. 2a shows NFS spectra of EuO at 69 GPa measured up to 130 K with $B_{\text{ext}} = 2\text{T}$, effecting a simple beat pattern in the completely magnetically aligned sample due to fm ordering. Above 130 K we measured without B_{ext} to determine the magnetic ordering temperature. Fig. 2b shows the derived values of S_{IS} , B_{hf} and T_{C} for EuO as function of the applied pressure in the NaCl and CsCl structure together with data from a previous study with conventional Mössbauer spectroscopy up to 31 GPa [3].

In the NaCl structure we extended the pressure range up to 44 GPa (coexistence of NaCl- and CsCl-phase) and found a strong increase of the isomer shift above 30 GPa towards trivalent Eu connected with a smaller pressure induced increase of B_{hf} and a further decrease of T_{C} . In the CsCl structure of EuO, the isomer shift indicates up to ≈ 70 GPa a divalent behaviour, similar to the observation for EuS and EuSe in that pressure region; the same holds for unusual low values of B_{hf} , caused by the different J_2 exchange which dominates the magnetic behaviour in the CsCl-type high-pressure phase [2]. Most interesting is the behaviour of T_{C} , where we observe for CsCl-type EuO first a strong increase of T_{C} up to 160 K at 78 GPa, and then, with the onset of (slightly) mixed-valent behaviour (as evidenced from the variation of S_{IS} and B_{hf}), a strong decrease of T_{C} to 110 K at 92 GPa. This behaviour is markedly different from EuS and EuSe and points to a reversal of the strength of the magnetic exchange interactions J_1 and J_2 in the Eu(II)-chalcogenides in their CsCl-structures [4].

References: [1] O. Leupold, K. Rupprecht, G. Wortmann, *Structural Chemistry* 14, 97-107 (2003). [2] K. Rupprecht, U. Ponkratz, O. Leupold, G. Wortmann, *ESRF Highlight* 2005, p. 10. [3] M.M. Abd-Elmeguid, R.D. Taylor, *Phys.Rev. B* 42, 1048 (1990). [4] K. Rupprecht et al., in preparation.