

**Experiment title:**

Dynamics of valence transitions in Eu compounds studied by nuclear scattering of Eu-151 21.5 keV-radiation. (report I)

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Report:

The present studies were performed in close collaboration with the Hamburg group (O. Leupold et al., HE-286) as well as with the Cologne group (M. Abd-Elmeguid et al., HE-175), see corresponding reports. Due to this special arrangement, the allocated shifts could be used very effectively. According to our proposal, the first high-pressure experiments with nuclear-forward scattering (NFS) of the 21.5 keV radiation of Eu-151 were performed. Here we report on pressure and temperature induced valence changes in intermetallic EuNi_2X_2 ($\text{X} = \text{Ge}, \text{Si}, \text{P}$) systems; these studies applied for the first time a new and effective method [1] to measure changes in isomer shifts (IS) by using additional reference absorbers. In Report II we present our high-pressure experiments on EuTe, where we probed Eu magnetism at various pressures and temperatures.

1. Pressure-induced valence transition from Eu^{2+} to Eu^{3+} in the intermetallic EuNi_2Ge_2 system at various pressures up to 12 GPa using a specially developed large-area high-pressure cell employing B_4C anvils with 1 mm absorber diameter. In these measurements we used EuS and EuF_3 as reference systems for the relative isomer shift determination. In Fig. 1(a) we show the NFS spectra of these absorbers measured alone and together. In the latter spectrum we observe a strong beating with a period of 4.8 ns corresponding to the 208 MHz energy difference in isomer shift between EuS and EuF_3 . The additional weak structure on the beating amplitudes is due to the small quadrupole splitting in EuF_3 , which is not resolvable in normal Eu-151 Mössbauer spectroscopy. Fig. 1 b shows the high-pressure NFS spectra of EuNi_2Ge_2 , indicating an almost complete valence transition from Eu^{2+} (0 GPa) to Eu^{3+} (12 GPa). Around 4 to 5 GPa the more complex spectra indicate a mixed-valent region with a distribution of valence states [2].

2. Temperature-induced valence transitions in the pseudo-ternary $\text{EuNi}_2\text{Ge}_{1.5}\text{Si}_{0.5}$ and valence changes in the homogenous mixed-valent EuNi_2P_2 system. These studies are of interest for the dynamical properties in the mixed valent systems and for comparison with the inhomogenous mixed-valent Eu_3S_4 system studied by the Hamburg group, which performed also first inelastic experiments (see report HE-286) demonstrating the feasibility of local phonon density-of-state spectroscopy with the Eu-151 resonance. Since these experiments are at present extremely time consuming, we postponed our proposed inelastic experiments on mixed-valent systems and performed only NFS experiments, where we measured at various temperatures 40 spectra at ambient pressure (taking 15 to 30 min each) and 28 spectra at high-pressure (taking about 1 h).

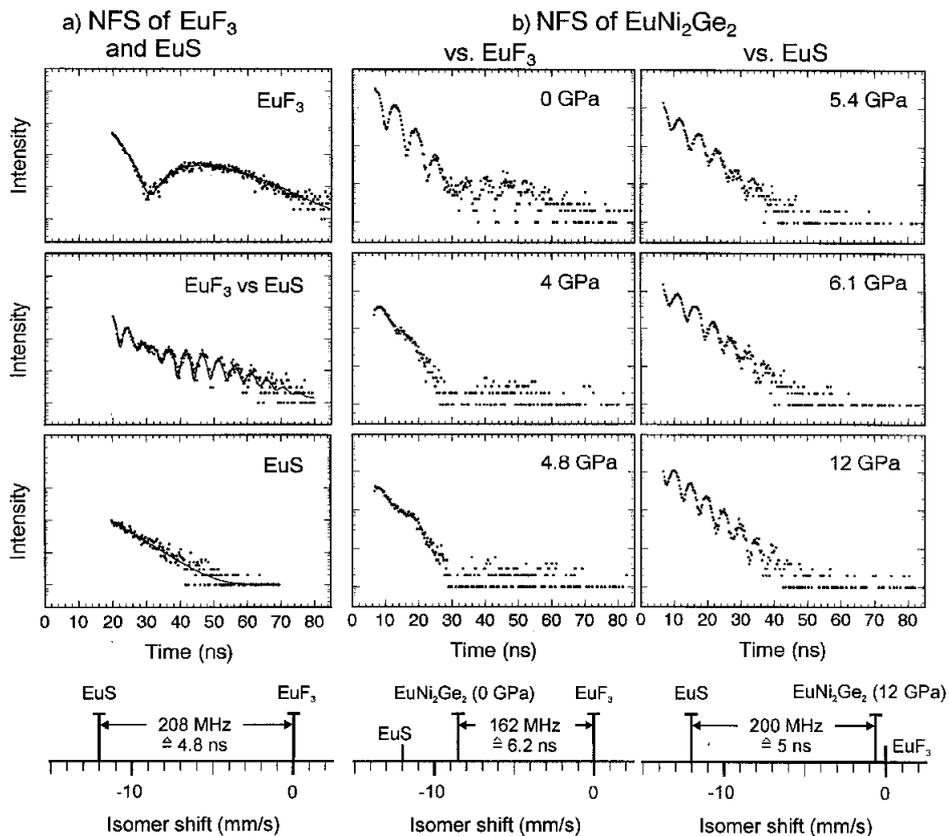


Fig. 1: (a) Eu-151 NFS spectra of EuF_3 , $\text{EuF}_3 + \text{EuS}$, and EuS . The pronounced minimum in the EuF_3 spectrum is due to thickness effects, the pronounced beating in the ($\text{EuF}_3 + \text{EuS}$) spectrum due to the difference in isomer shift, slightly modified by the quadrupole interaction in EuF_3 . (b) Eu-151 NFS spectra of EuNi_2Ge_2 at various pressures measured together with EuF_3 (left column) and EuS (right column) as reference absorbers. The observed beatings in the time spectra are due to the differences in isomer shift between EuNi_2Ge_2 and the reference absorber (see bar diagram below).

References:

- [1] O. Leupold et al., Europhys. Lett. 35, 671 (1996).
- [2] H.-J. Hesse et al., J. Alloys Compounds 246, 220 (1997).