ESRF	**Experiment title:** Measuring the electron hopping frequency in valence mixed iron of the RBaFe$_2$O$_5$ double perovskite with (R = Gd and Pr)	**Experiment number:** HC4024
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Report:

RBaFe$_2$O$_5$ (R =Y, Ho, Dy, Gd, Sm, Eu, Nd or Pr) is a double-cell perovskite that exhibits valence-mixing (Fe$^{2.5+}$) at high temperatures and charge-ordering of Fe^{2+} and Fe^{3+} at low temperatures.[1] At the Verwey temperature T_V the charge-ordered phase undergoes a transition to an intermediate phase with partially separated valences of Fe$^{2+\epsilon}$ and Fe$^{3-\epsilon}$ and finally at a second transition temperature $T_p > T_\mathrm{V}$ a full mixing is achieved. The full and partial mixing is connected to a ferromagnetic coupling along the c-axis through the basal planes of the cooridnation pyramids of the Fe species. Below T_V the Fe atoms couple antiferromagnetically with their nearest Fe neighbors. The aim of this project was to see if the dynamics of the mixed, intermediate and mixed states could be observed using nuclear forward scattering on two double-perovskite powder samples based on Gd and Pr, having differing transition temperatures T_p, T_V. Previously obtained spectra using traditional energy-domain (ED) Mössbauer spectroscopy reveal clearly the six types of Fe species found in the three temperature regions of $T < T_\mathrm{V}$, $T_\mathrm{V} < T < T_p$, and $T_p < T$ at the two lattice postions of the unit cell.

A total of 46 NFS spectra measured for 15-45 minutes were gathered for both samples at several temperatures between 5 and 317 K, thus covering all three temperature regions. Upon passing the transition temperatures the data changed profoundly, as seen also in ED measurements. All six iron species can be identified from the NFS data and internal field values, qaudrupole coupling constants, as well as isomer shifts agree with the ED data. We also measured a few spectra in external fields of 1 and 4 T and at temperatures represenative of the valence-mixed state, the intermediate

state and the charge-ordered state (Gd only).
Preliminary results seem to indicate that the charge-ordered state is partially forced into the intermediate state by the external field, possibly by favouring the local ferromagnetic coupling along the *c*-axis. During the measurements the forward scattered count rate was carefully monitored. It turned out that upon passing T_V a strong decrease of the rate was observed and the rate remained low until T_p was reached when the rate practically recovered to its low-temperature value again. No such observation have been seen in data obtained in the ED. Tentatively the dip in count rate has been assigned to loss of coherence caused by a broadening of the resonance lines (also seen in ED) within the intermediate state. This is supported by the fact that the external field caused a similar dip in the count rate for all recorded spectra, as an external field clearly causes magnetic broadening in an overall antiferromagnetic order.

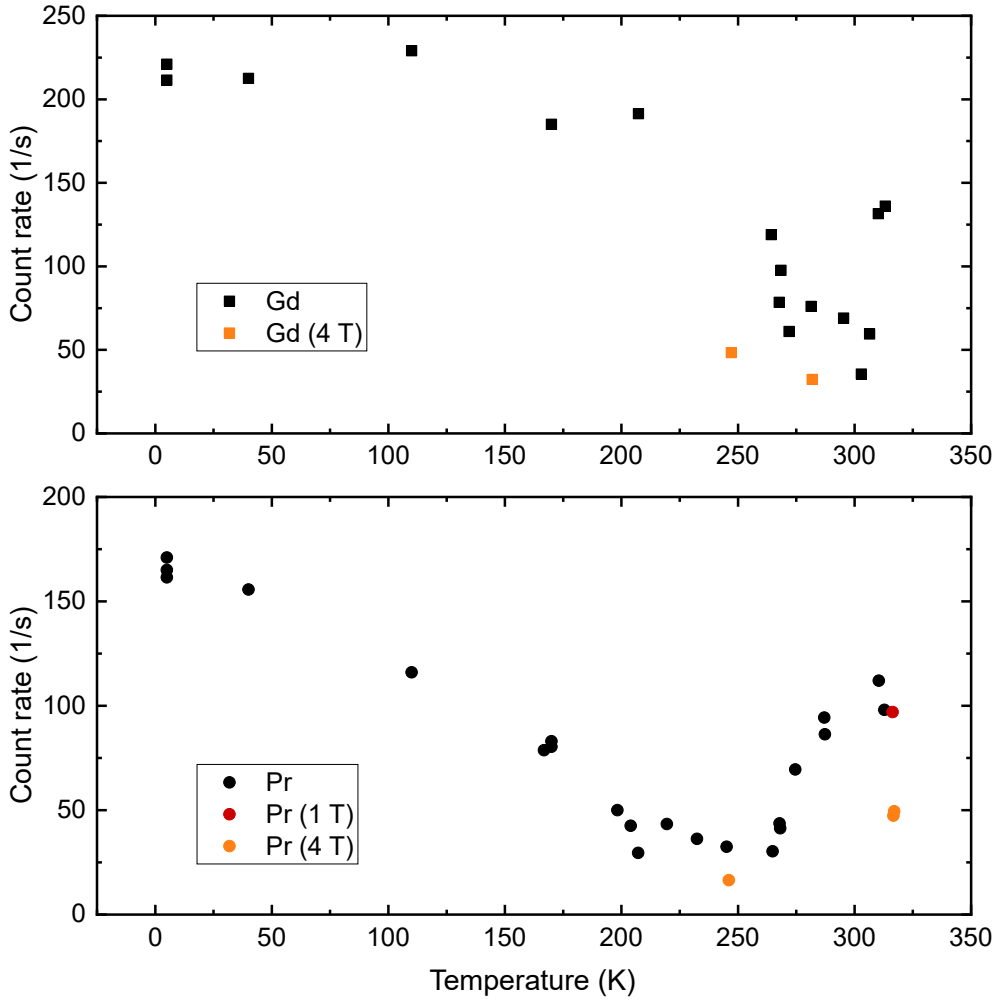

Fig. 1 Experimental count rates in the NFS measurements.

[1] J. Lindén, F. Lindroos, and P. Karen, J. Solid State Chem.**252**, 119-128 (2017).