ESRF	**Experiment title:** High Q_{max} synchrotron powder diffraction data for high resolution atomic pair distribution functions of icosahedral Mg-Zn-*RE* quasicrystals (*RE* = Ho; Y)	**Experiment number**: HS-2629
Beamline: ID31	**Date of experiment**: from: 17-sept-04 to: 21-sept-04	**Date of report**: 21-jan-05
Shifts: 12	**Local contact(s)**: Michela BRUNELLI	*Received at ESRF:*

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Experiment HS-2629 refers to proposal Ref. 7505.

Report:

The 3D real space atomic structure of quasicrystals is still no straightforward task to solve. Within the family of icosahedral (*i*) quasicrystals, some Mg-Zn-*RE* phases (*RE* = Y and various rare earths; *fci*: *Fm*-3-5 and *si*: *Pm*-3-5) are distinguished from others by a minimum structural disorder as indicated by the lack of diffuse scattering in diffraction experiments [1, 2]. This predestines them for structural investigations:
A new complementary approach is to derive their local structures *via* the atomic pair distribution function (PDF or $G(r)$; [3]) from powder diffraction data [4].

The purpose of experiment HS-2629 was to check the recent local quasicrystal structure analyses using PDF data from in-house X-ray powder diffraction data (Q_{max} = 13.5 Å^{-1}) of *fci*-$Ho_9Mg_{26}Zn_{65}$ and *si*-$Ho_{11}Mg_{15}Zn_{74}$ [4, 5], produce more accurate data and yield accurate data for *fci*-$Mg_{25}Y_{11}Zn_{64}$.

The success of the experiment is documented in a paper "PDF from X-ray powder diffraction for nanometer-scale atomic structure analysis of quasicrystalline alloys" which has been submitted to *Z. Kristallogr.* [6];
Abstract: *For three icosahedral alloys in the Mg-Zn-*RE *(*RE *= Ho, Y) systems, the atomic pair distribution functions (PDFs) have been obtained from sealed X-ray tube (in-house) and synchrotron (ESRF and APS) powder diffraction experiments. The PDFs are at least qualitatively comparable and are suitable for least squares $r \approx 2$ nm local structure refinements. A recent local model for si-Ho-Mg-Zn from in-house data can be confirmed using the synchrotron data. Acquisition and the quality of the data from the different sources are discussed. The better resolved synchrotron data open perspectives for larger scale and thus more detailed models.*

To achieve a $Q_{max} > 30$ Å^{-1}, the wavelength $\lambda = 0.30235(1)$ Å was selected at ID31 and the measurements were performed at the diffraction range $0° < 2\theta < 120°$.

The quality of the PDF as Fourier transform of the normalized structure function from powder diffraction data is dependent on good counting statistics at high Q [3]. Therefore the time dedicated to a step in Q in the 2θ-scans was tripled above $Q > 26$ Å^{-1}. Special care has to be taken about a possible joint between the different scan velocities. Only proper normalisation w.r.t. monitor counts and a background subtraction of an empty sample measurement (same scan parameters!) will avoid disturbing effects in the Fourier transform ("chequered" PDF). Summation of a minimum of 10 scans (~10 h measuring time) yields very well-resolved diffractograms. Binning to $\Delta 2\theta = 0.01°$ proved to be sufficient for PDF generation using the program PDFgetX [7].
This measuring mode results from optimisation using the *fci*-$Mg_{25}Y_{11}Zn_{64}$ sample first as it was the alloy with lowest Z elements (and thus lowest X-ray contrast) of the quasicrystals investigated. Figure 1 (a-c) show the well-resolved PDFs for the three samples. Termination effects decrease with increasing Z of the samples.

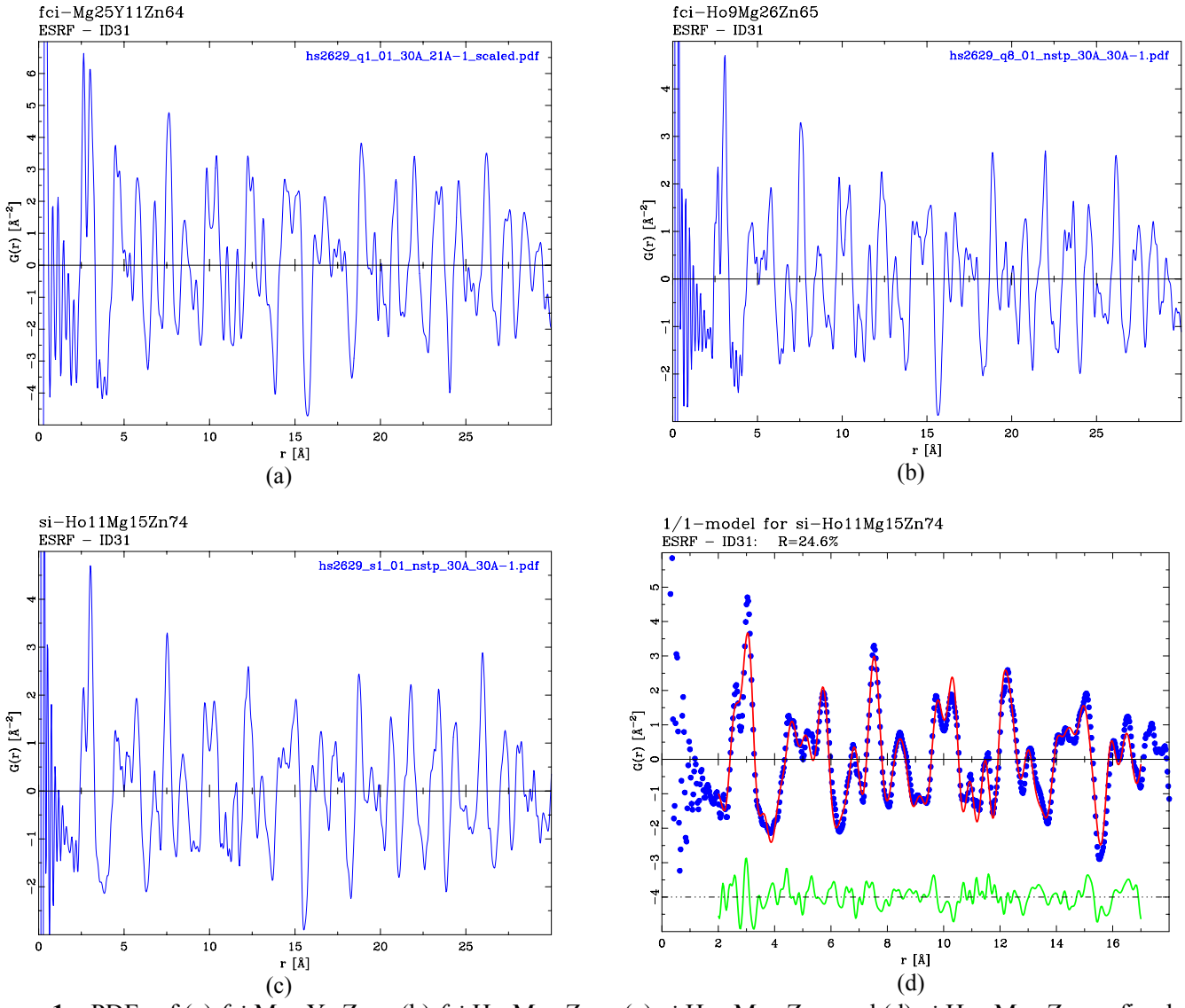

Figure 1. PDFs of (a) *fci*-$Mg_{25}Y_{11}Zn_{64}$, (b) *fci*-$Ho_9Mg_{26}Zn_{65}$, (c) *si*-$Ho_{11}Mg_{15}Zn_{74}$ and (d) *si*-$Ho_{11}Mg_{15}Zn_{74}$ refined as 1/1 virtual approximant model ($R = 24.6\%$) [5] – dots: experimental PDF, red continious line: calculated model PDF, green bottom curve: difference plot.

A least squares refinement [8] of a local 1/1-model using the PDF up to $r = 17$ Å is displayed in Figure 1(d) ($R = 24.6\%$). Qualitatively it confirms the refinements of the in-house data ($R = 20.4\%$) [5]. Besides a more reasonable scale factor with the synchrotron data [5], existing differences model-to-data can be seen now more clearly. They are expected to be minimized in larger approximant (*e.g.* 2/1) models. For the such future refinemetns, data from ID31 will serve as an excellent basis.

In [6] we compare the ESRF (ID31) data presented here to some *i*-Mg-Zn-*RE* data from APS (6ID-D; Argonne National Laboratory). The Image Plate setup and a rapid data acquisition technique with inferior *Q*-resolution as used there is more time efficient and yields good *r*-resolved PDFs. But longer measuring times with better resolution in *Q* at ID31 are rewarded by more clear information of $G(r)$ in high *r* regions. This is needed for larger scale "local" models.

Another intermetallic sample, the alloy β-Al_3Mg_2 (cF1832-664, *Fd*-3*m*, $a = 28.34$ Å [9]; courtesy of M. Feuerbacher, FZ Jülich, Germany), was measured during HS-2629 at ID31. It represents one of the most *complex metallic alloys* (CMA) in terms of unit cell dimensions and atomic disorder [10]. As the PDF comprises a local picture of intrinsic disorder of materials [3], we plan to investigate β-Al_3Mg_2 using the PDF as a local structure probe.

Figure 2 shows a fairly spiked PDF of β-Al_3Mg_2, obtained from ID31 data with the same measuring parameters as used for *i*-Mg-Zn-*RE*. Note the large truncation oscillations for $r < 2$ Å. The low *Z* of the materials components should be accounted for by optimised measuring conditions and time.

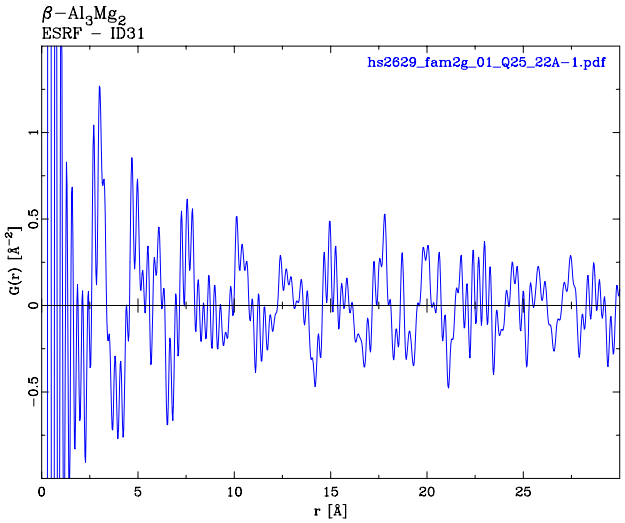

Figure 2. PDF of β-Al_3Mg_2, the quality of the data should be improved by optimised measuring conditions

Conclusion:

- PDFs from ESRF – ID31 ($Q_{max} = 30$ Å^{-1}) data are well resolved in *r* and have a reasonable scaling. They show a very detailed picture of the PDF peaks and will serve better for larger scale models.
- Good resolution in *Q* at ID31 ensures good distinction of PDF peaks at high *r*. This is an advantage over time-saving data acquisition using an Image Plate *e.g.* at APS.
- The 1/1 local structure model for *si*-Ho-Mg-Zn, *i.e.* the topology, geometry and element distribution of its basic cluster [5], can be confirmed using the data from ID31 [6]. Nevertheless, the new data point at local inconsistencies of the rather small 1/1-model with the "true" quasicrystal structure.
- PDF generation is more robust for higher-*Z* materials.

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