ESRF	Experiment title: **High pressure study of heavy-fermion superconductor PuXGa5 (X = Co, Rh, Ir)**	Experiment number: **HS 2338**
Beamline: **ID30**	Date of experiment: **07 July 2004 to 12 July 2004**	Date of report: **9/02/05**
Shifts: **15**	Local contact(s)**: Dr Mohamed MEZOUAR**	*Received at ESRF:*

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Report:

High-pressure, angular-dispersive, X-ray powder diffraction measurements have been performed on the heavy-fermion superconductors $CeXIn_5$ (X = Co, Rh) and $PuCoGa_5$. The main interest in this work was the evolution, as a function of pressure, of the *c/a* ratio in each of these tetragonal (P4/mmm) systems. This followed recent evidence for structural tuning of the superconductivity in these (Ce, Pu) '115' families of compound [1]. It is worth noting here that our original proposal to ID30 planned to investigate several compositions from the Pu 115 family (i.e. $PuXGa_5$ for X = Co, Rh and Ir), *only*. However, within the intermediate period, between proposal submission and experimental preparations (i.e. sample-DAC loadings), we became aware of recent high-pressure diffraction studies on Ce 115s made at the APS [2,3], which, for $CeRhIn_5$ in particular, concluded a double-peak dependence of *c/a* on pressure ($0 < P < 6$ GPa), a dependence which, we believe, is *not* strongly supported by the data in ref. 2. On this basis, we decided to study only one Pu composition, i.e. $PuCoGa_5$ (X = Co), and devote the remaining beamtime to (re-)investigations of compositions from the Ce 115 family; in particular, to confirm, or otherwise, the double-peak *c/a*(P) for $CeRhIn_5$, using *superior* pressure conditions (i.e. greater hydrostaticity) to those achieved in ref. 2.

Powder samples of $CeXIn_5$ (X = Co, Rh) and $PuCoGa_5$, each of a similar quantity (~ 10 μg), were loaded into diamond-anvil cells of the Syassen-Holzapfel type ($2\theta_{max}$ ~ 14 deg.), with diamond culets of 300 μm (diameter) and fitted with inconel gaskets, each with a 150 μm (diameter) hole. In the case of the Pu compound, to avoid the risk of contamination, the powder was combined with epoxy resin ('glue') to form a solid piece, prior to loading. Silicon oil was used as the pressure transmitting medium (PTM) for all compounds, except $CeXIn_5$ where two loadings were made, one with oil and the other with nitrogen (both, especially the latter PTM, constitute improved hydrostaticity as compared to the studies of ref. 2). Using a collimated (40 μm × 40 μm) beam of a 61.33 keV (0.20215 Å) X-rays (from ID30), and calibrating pressure using the ruby-fluorescence method, diffraction patterns were collected (using a MAR345 image-plate detector) for each compound, at pressure increments of ~ 0.5 to 1 GPa, up to P_{max} ~ 15 GPa for $CeXIn_5$ and ~ 37 GPa for $PuCoGa_5$. These patterns were then processed using ESRF software (Fit2D) to obtain a data-format suitable for structural analysis. Since the main interest in this work was the pressure evolution of the *c/a* ratio, structural analysis of diffraction patterns was limited to lattice parameter determination, only, using the *profile-matching* (or 'Le Bail') technique [4] implemented using the program *Fullprof.* Examples of the quality of data fits generally achieved are given in Fig. 1.

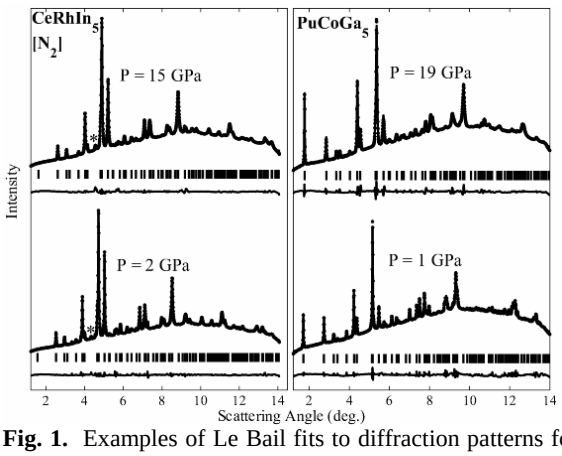

Fig. 1. Examples of Le Bail fits to diffraction patterns for Ce and Pu 115s, taken at several pressures.

The *c/a*(P) results for $CeRhIn_5$ are in discord with the double-peak variation deduced in the previous work (ref. 2); our observation is that of a monotonic increase in *c/a* with pressure, up to P = 8 GPa, as shown in Fig. 2(a) (upper panel). A comparison between these results and those from ref. 2 is also shown (lower panel). We observe variations in *c/a* (with each P step) that are far more uniform than those in ref. 2, the points from this earlier study being 'scattered' about dashed curve (lower panel). We therefore argue that our measurements represent an improvement on these earlier studies. We argue likewise for $CeCoIn_5$,

however here we obtain at least some agreement with earlier work (ref. 3), in that, in the range 0 < P < 15 GPa, *c/a*(P) is a single-peak function, with peak position between 3 and 5 GPa, as shown in Fig. 2(b), where (again) the results from the previous study (ref. 3) are reproduced, for comparison. In addition, we present in Fig. 2(b) (top-panel) previously unpublished values of *c/a*(P) for $CeCoIn_5$ for P < 5 GPa (open-squares), obtained in an ID30 study carried out several years ago [5]. These data, collected under similar experimental conditions to ours (i.e. powder sample quantity and PTM) are in excellent agreement with the trend we observe. In Fig. 2(c) we show the 'compressibility curves' for each Ce 115 compound, i.e. the P dependence of V/V_0 (= $(a^2c)/(a^2_0c_0)$). The fits are to a Birch-Murnaghan (B-M) form for the equation of state (EOS). The fitted value of Bulk modulus (B_0) for $CeCoIn_5$ is 76.1(14) GPa, in good agreement with that of 78.2(18) GPa previously reported [3], whereas our B_0 for $CeRhIn_5$, at 85.4(11) GPa, is higher than the value of 78.4(2) GPa previously measured [2]. However, as with the trends in *c/a*(P) for $CeRhIn_5$, our measured values of V/V_0(P) show less deviations ('scatter') about the fitting curve and we thus conclude that our B_0 (for $CeRhIn_5$) is more reliable than the value in ref. 2 (this also follows from the superior hydrostaticity which will have been achieved in our studies).

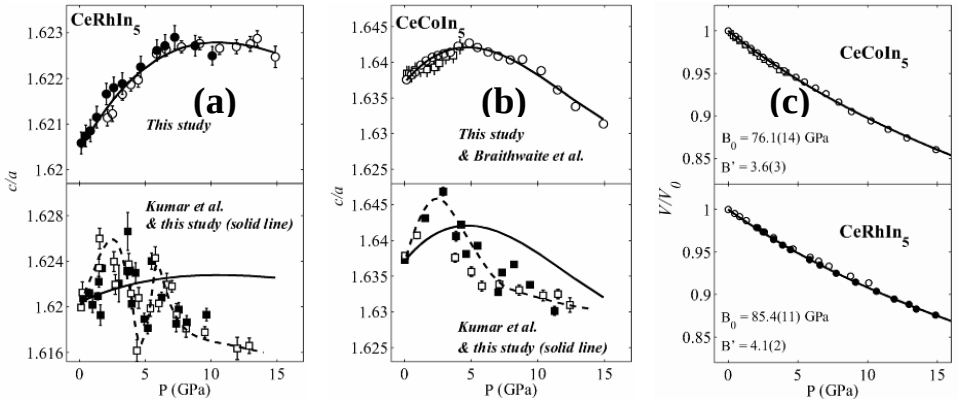

Fig. 2. *c/a*(P) for, **(a)**, CeRhIn5 and, **(b)**, CeCoIn5. In **(a)** and **(b)**, the upper panels show our results and the lower panels compare these results (solid lines) with results (points and deduced trends (dashed lines)) from refs. 2 and 3. Compressibility curves for both compounds are in **(c)**.

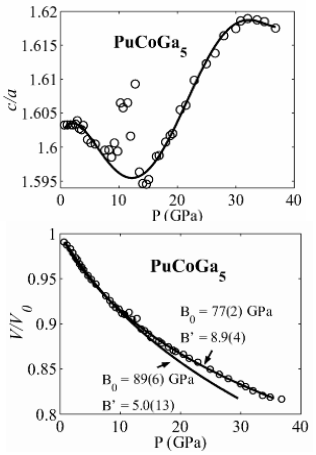

Fig. 3. *c/a*(P) (upper panel) and compressibility curve (lower panel) for $PuCoGa_5$.

For PuCoGa5, we deduce that *c/a*(P) is, in the range 0 < P < 37 GPa, a function with a single minimum at position 12-13 GPa, as shown in upper panel of Fig. 3. However, in the range 10-13 GPa we observe an anomalous behaviour (a 'relaxation' [6]) in the *c* lattice parameter which results in the deviation of the (*c/a*) points (Fig. 3(upper panel)) from the 'deduced' trend (solid line). We believe that this *c* parameter anomaly is associated to the known 'freezing' of silicon oil (PTM) seen to occur ~ 8 GPa [7]. That such an effect is not observed in any of our experiments (using oil as PTM) on Ce 115s is, we believe, due to slight differences in the experimental conditions between the Ce and Pu 115 studies; the crystallites (powder grains) of PuCoGa5 were larger and more *plate-like* than those in the Ce 115 studies, and, in addition, were bound together by glue [6]. The lower panel in Fig. 3 shows the corresponding compressibility curve for $PuCoGa_5$. Again, we observe an anomaly at ~ 10 GPa, but this time it is a much weaker effect than that seen in *c/a* (at ~ 10 GPa, only two data points in Fig. 3 (lower panel) deviate from the B_0 = 77(2) GPa fitting curve). This can be understood, as the unit-cell volume ($V = a^2c$) is less sensitive to variations in *c* than the quantity *c/a*. Two B-M EOS fits are shown in the figure. The fit corresponding to the larger B_0 was made with data for P < 10 GPa, only, whereas the second fit took into account the full presesure range. We believe that due to the anomaly in the *c* parameter, one cannot have confidence, quantitatively, in *V*(P) beyond 10 GPa (whereas, qualitatively, we may still conclude that *c/a* increases for P > 13 GPa). Thus, we believe 89(6) GPa to be the more reliable deduction of B_0 in this system (this fit also gives a value of B' (at ~ 5) more typical of actinide intermetallic systems).

In Fig. 4, we plot the derived trends (from the diffraction data) in *c/a*(P) for all compounds investigated [8], and compare these with the *c/a*(P) ('anticipated') which would be expected if, as suggested in ref. 1, T_c(P) were correlated to changes in *c/a* occurring under pressure [9]. From the disagreement with expectation, for all systems, we conclude the absence of such (simple) correlation.

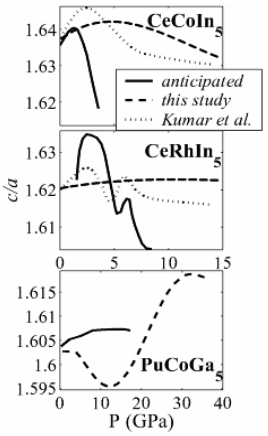

Fig. 4. Summary of results showing incompatibility of all measured *c/a*(P) trends with the anticipation that *c/a* acts as a superconductivity tuning-parameter (see ref. 1)

References and Notes:

[1] Bauer et al., Phys. Rev. Lett. **93**, 147005 (2004).
[2] Kumar et al., Phys. Rev. B **69**, 014515 (2004).
[3] Kumar et al., Phys. Rev. B **70**, 214526 (2004).
[4] LeBail *et al.*, *Mat. Res. Bull.* **23**, 447 (1988).
[5] Braithwaite, Le Bihan, Mezouar, unpublished results (ID30, July 2002).
[6] A more detailed account of this work is to be submitted to Phys. Rev. B.
[7] See, e.g., Le Bihan et al., Phys. Rev. B **67**, 134102 (2003).
[8] We also plot the derived trends from refs. 2 and 3 (Ce 115s). For $CeRhIn_5$, we observe that although the peak positions in the trend agree with anticipation, the variations ($\Delta(c/a)$) do not.
[9] Each 'anticipated' curve was obtained by multiplying the measured T_c(P) by the value of $(dT_c/d(c/a))^{-1}$ for the 115 family (see ref. 1), and then shifting the resulting curve to the correct c_0/a_0 value.