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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.

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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.

ESRF	**Experiment title:** δ-phase stabilization in $Pu_{(x)}Am_{(1-x)}$ alloys as studied by EXAFS	**Experiment number**: HE1393
Beamline: BM 20	**Date of experiment**: from: 30 January 2003 to: 02 February 2003	**Date of report**: 5 august 03
Shifts: 12	**Local contact(s)**: C. Hennig	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):
RAVAT, JOLLY, VALOT, BACLET
CEA-Centre de Valduc

Report:

The plutonium δ-phase (face-centered cubic) can be stabilized at room temperature with so-called deltagen elements such as Ga, Al, Am, Ce… While PuGa and PuAl alloys have been extensively studied, few works have been devoted to PuCe and PuAm alloys. A Ph.D. (initiated in 10/98) was devoted to a better understanding of the δ-phase stabilization in especially binary $Pu_{(x)}Am_{(1-x)}$ alloys [1]. Electronic and crystalline structure appeared to be strongly related in these alloys. Electronic effects have been suggested through a positive deviation from the Vegard's law, as shown by XRD [2]. A strong localization of the 5f electrons of Pu atoms has been indicated by electrical and magnetic measurements for an Am content of about 25 at% [1]. However, the techniques used until now gave only an overall information, whereas, as EXAFS measurements in $Pu_{(x)}Ga_{(1-x)}$ alloys revealed, important phenomena can happen at a short range order [3,4].

Experiment

EXAFS measurements have been performed at the LIII-edges of Pu (18057 eV) and Am (18504 eV) in the transmission mode on four alloys: $Pu_{0.95}Am_{0.05}$, $Pu_{0.85}Am_{0.15}$, $Pu_{0.75}Am_{0.25}$ and $Pu_{0.57}Am_{0.43}$. EXAFS spectra have been recorded at 20 K using the cryostat from the BM20 line.

Results and discussion

Preliminary theoretical study

The LIII-edge of Am being close to Pu one, the EXAFS spectrum of the Pu interferes with the Am one. Therefore, using FEFF calculation, the theoretical FT of the Pu, Am and PuAm alloy EXAFS spectra have been compared. The results (Fig.1) show that the Am-Pu contribution is not influenced by the Pu signal. The addition

of the Pu signal does not modify the peak position of the Am-Pu contribution. A study of the Am-Pu distances is then possible by filtering this contribution in experimental spectra.

Experimental study

The results (Fig. 2) highlight an increase in both Am-Pu and Pu-Pu distances with the Am content and the Am-Pu distances are always larger than the Pu-Pu ones with an expansion of 0.03Å. Moreover, an increase in cell parameter with Am content has been equally observed by XRD, technique which is sensitive to long range order. The values obtained by XRD tend towards the Pu-Pu distances for the lowest Am content and towards the Am-Pu ones for the highest Am content. This can first be attributed to a steric effect, the Am atomic volume being more important than the Pu one.

However, as the Am content increases, Pu atoms are replaced by Am atoms whose atomic volume is larger. Consequently, the resulting lattice expansion induces a decrease in the overlapping of the 5f wave function of the plutonium. This phenomenon leads to an increase in the 5f electrons localization and thus an increase in the atomic volume. So, the increase in the Am-Pu and Pu-Pu distances are the result of both steric and electronic effects.

It is noticeable that Pu atoms seem to reach their 5f electrons maximum localization degree in this kind of alloys, as it was suggested by the susceptibility result [1], since a slight decrease in the slope can be observed from an Am content of 25%. This effect, only observed for the Pu-Pu distances, shows that, from this Am content, only the steric effect remains. Finally, the great difference between the Am-Pu and Pu-Pu distances for an Am content of 43% seems to highlight that the distances for both pairs will be never equal whatever Am content. This involves that the 5f e^- localization degree of Pu could not reach the 5f electron localization degree of Am in this alloy.

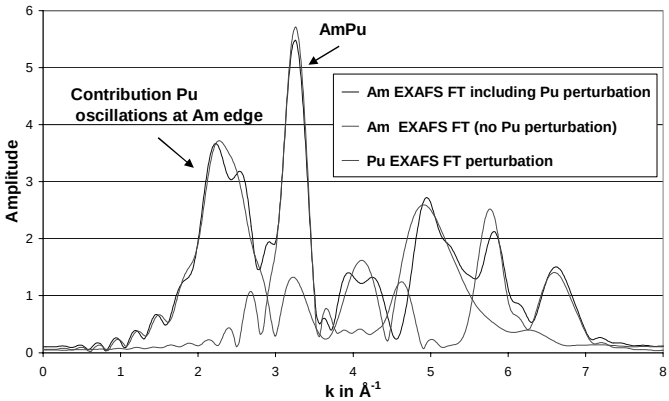

Fig. 1: Theoretical EXAFS FT calculation using FEFF7.

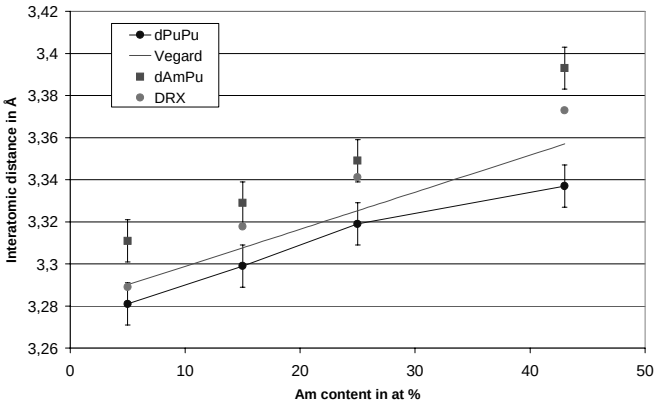

Fig. 2: Interatomic distances for the Am-Pu and Pu-Pu pairs.

Acknowledgement

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[1] M. Dormeval, “Structure électronique d’alliages Pu-Ce(-Ga) et Pu-Am(-Ga) stabilisés en phase δ”, thèse de l'Université de Bourgogne, septembre 2001.
[2] M. Dormeval, N. Baclet, "Plutonium Futures - the Science", july 2000, Santa-Fe, New Mexico.
[3] Ph. Faure, B. Deslandes, D. Bazin, C. Tailland, J.M. Fournier, J. Alloys and Comp., 244, p131 (1996).
[4] S. D. Conradson,“Where is the Gallium?”, Los Alamos Science Number 26, 2000.