



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.

Published papers

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.



	Experiment title: Electron density study of hydrogen bonding in NH ₄ F	Experiment number: HS1741
Beamline: ID15B	Date of experiment: from: 26.06.02 to: 02.07.02	Date of report: 29.08.02
Shifts: 18	Local contact(s): Nozomu HIRAOKA	<i>Received at ESRF:</i>
Names and affiliations of applicants (* indicates experimentalists): SHUKLA Abhay* BELLIN Christophe* AMEIL Cédric* BUSLAPS Thomas*		

Report:

The present experiment takes place within a particular aim of our group which is to study hydrogen bond through a large spectrum of systems such as urea [1], ice I_h [2], NH₄F [present work] or water (proposal deposited). The NH₄F experiment follows directly the one performed on ice I_h since both materials are important examples of cooperative systems containing moderately strong hydrogen bond. Moreover, NH₄F is isomorphic to ice with an hexagonal structure in the polar space group P6₃mc and hydrogen bonds are weaker in this system than in ice.

The experiment has been performed with the scattering angle settled at 173 degrees and the synchrotron radiation has been monochromatized to 29 keV. Crystals have been oriented in such a way that one could measure Compton profiles along *a* and *c* axis.

The data sets have been corrected for energy dependent effects such as photoelectric absorption in sample, analyser and air (photon path between sample, analyser and detector). Since the only electrons of interest are valence electrons, calculated core Compton profile has been subtracted from the total profiles using QSCF approximation [3]. The resulting valence profiles were then normalized to the number of valence electrons, and corrected from the multiple scattering contribution.

In order to examine the effects of hydrogen bonding, we first have measured anisotropies ($J_{c\text{axis}} - J_{a\text{axis}}$): this difference of measured Compton profiles is virtually free from systematic errors. Moreover, what remains in the anisotropy is dominated by wave function phase coherence among neighboring molecules through the hydrogen bond. The second way to analyse experimental results is to take the power spectrum of the anisotropy defined above: it allows us to come back to real space and get information on coherent electronic interaction in real space which may modulate Compton profile in momentum space.

Both ice and NH₄F globally exhibit same features and modulations in their anisotropies. The difference in amplitude may result from the more isotropic character of NH₄F, which in other terms reflects a weaker

hydrogen bond. We notice that both anisotropies differ in the low momentum part which carries the information on the more delocalized electrons, thus in particular hydrogen electron.

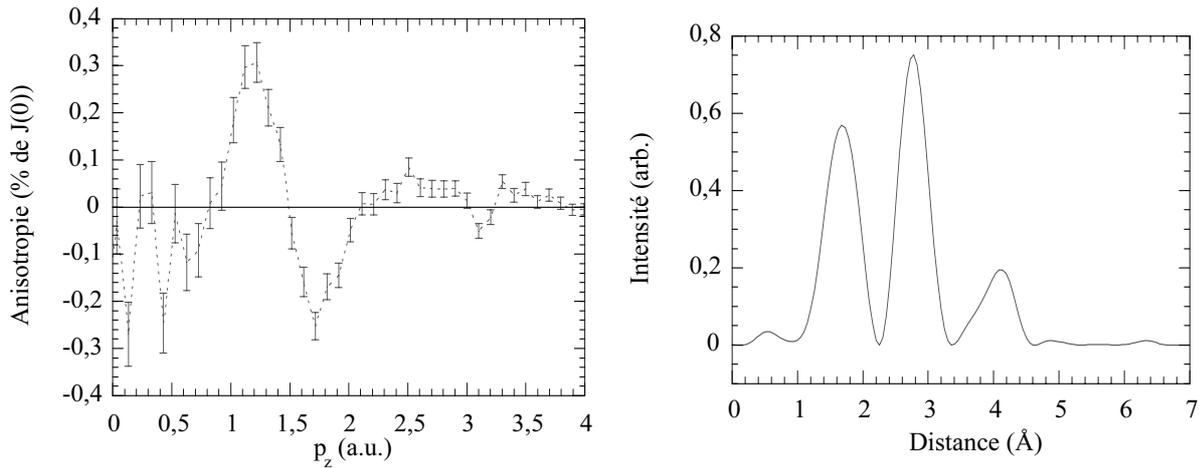


Figure 1- on the left : anisotropy between profiles measured along c and a axis with counting statistics error bars; on the right : power spectrum of the measured anisotropy

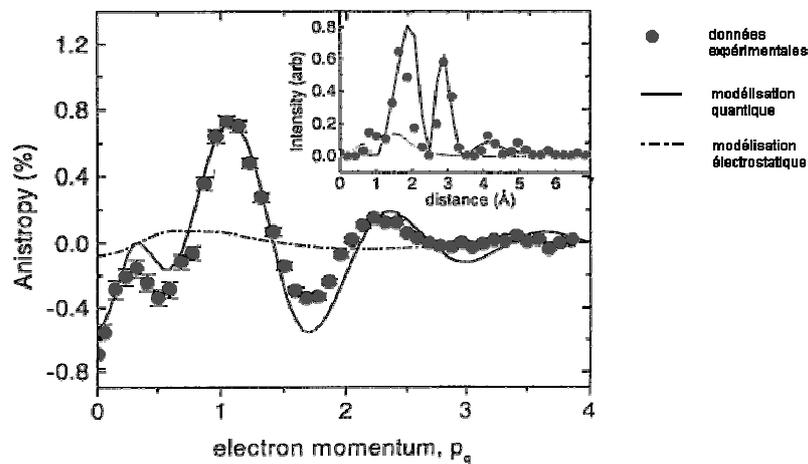


Figure 2 - anisotropy measured on ice between profiles measured along c and a axis with counting statistic errors [2]. In the quadrant : power spectrum of the ice measured anisotropy.

We compare both power spectrum obtained in ice and NH_4F . S. Ragot and coworkers [4] shown that the first peak at 1.6 \AA is due to electronic correlations. In the case of ice, ED Isaacs and coworkers attributed the second and third peaks to real space distances respectively between two oxygen atoms and between oxygen and hydrogen in a H-bond. What is observed in NH_4F is that the second peak may reflect the $\text{F}\cdots\text{H}$ distance while third and fourth peaks are roughly at the same positions that in ice.

B. Barbiellini is currently performing new calculations on hydrogen bond. They will be applied to NH_4F and thus will help us both to analyse NH_4F results alone and to compare results obtained for ice and NH_4F . From such a comparison should arise a more detailed understanding about the charge transfer within hydrogen bond.

References :

- [1] A. Shukla, ED Isaacs, DR Hamann, PM Platzman, Phys. Rev. B **64**, 052101 (2001)
- [2] ED Isaacs, A. Shukla, PM Platzman, DR Hamann, B. Barbiellini, CA Tulk, Phys. Rev. Lett **82**, 600 (1999)
- [3] A. Issolah, Y. Garreau, B. Lévy, G. Loupiau, Phys. Rev. B **44**, 11029 (1991)
- [4] S. Ragot, JM Gillet, P. Becker, Phys. Rev. B **48**, 235115 (2002)