

Beamline: ID15B	Date of experiment: from: 23/04/2003 to: 01/05/2003	Date of report: 26/02/2004
Shifts: 21	Local contact(s): N. Hiraoka	<i>Received at ESRF:</i>
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Report:

LiH is the simplest heteronuclear compound. Yet it still poses a challenge to theoretical chemistry mainly due to the highly diffuse character of the H^{-1} ions. On the other hand it is a good candidate for inelastic x-ray scattering (IXS) studies because it consists of low- Z elements and it possesses weakly bound valence shell electrons, making it an attractive target for Compton scattering studies. In spite of its simplicity the exact electronic properties of LiH are still subject to controversy even today. In this experiment, we collected a large set of directional Compton profiles with good accuracy in order to reconstruct the electron density matrix, which can be compared to results of charge density studies.

The experiment was performed at the high-energy inelastic x-ray scattering beamline ID15B. We measured 15 directional Compton profiles of single-crystalline LiH samples using the scanning-crystal spectrometer. Incident photon energy was 57 keV. The spectrometer utilizes the Rowland circle geometry based on a Ge(440) cylindrically bent analyser crystal. The momentum space resolution was 0.2 atomic units (a.u.) with the dominating factor being the thickness of the samples. The statistical noise was about 0.1% at the Compton peak. The samples were in the shape of 15 mm long sticks with a 1 mm \times 1 mm square cross section, cut with the crystal faces cut along the (100) and (110) planes. The measured 15 crystal directions were chosen to sample the irreducible 1/48th zone of the crystallographic unit cell as densely as possible.

Naturally when turning the sample to match the scattering vector along these directions, the effective thickness of the sample and thus also the resolution of the spectrometer changes. We considered the change in the resolution function by calculating the exact resolution in each case of sample geometry. The resulting resolution functions had full-width-at-half-maximums ranging from 0.199 a.u. to 0.218 a.u. These changes were considered in the data analysis by convoluting all theoretical Compton profiles with the corresponding experimental resolution function.

An example of our results is shown in Figure 1, which depicts the differences between directional [110] and [100] LiH Compton profiles and the spherically averaged profile (i.e. the anisotropic term). In the figure we also report the first results of our calculations for theoretical Compton profiles. The Hartree-Fock calculations (solid lines) were performed using the *ab-initio* program CRYSTAL98. We also used cluster calculations where only the exchange interaction was considered (dashed line) to separate the effects of polarization and charge transfer in the bonding. The rather good agreement between the Hartree-Fock calculation and the experimental result suggests that correlation effects are small in this system.

Density-functional Compton profile calculations are being prepared, and the reconstruction of the three-dimensional momentum density and of the full density matrix are in progress.

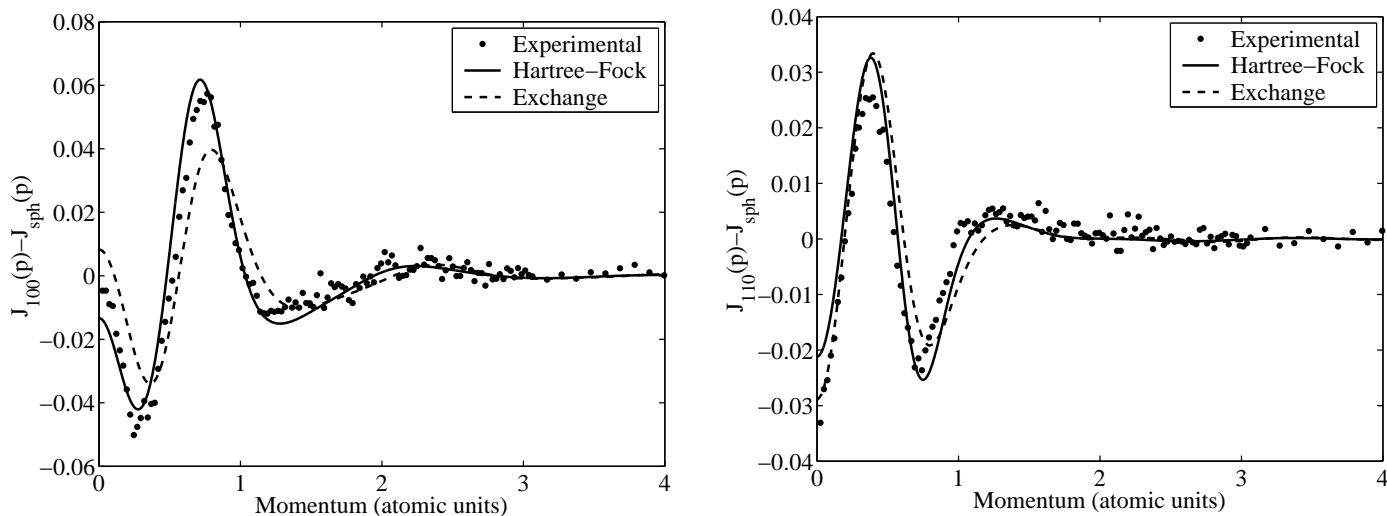


Figure 1. Left: Difference between the Compton profile along [100] and the spherical average. Right: Same as left, but between [110] and the spherical average. The dashed line presents the theoretical results after only performing Löwdin orthogonalisation of the ionic wave functions. The solid line is the theoretical anisotropy within the level of Hartree-Fock calculation.