Experimental electronic charge density of complex hydrides from maximum entropy method

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Alkali and alkali-earth complex hydrides have attracted growing interest as hydrogen storage materials thanks to their high gravimetric and volumetric density. However most of the alkali complex hydrides only desorb hydrogen after melting, due to either the thermodynamical stability or a kinetic barrier. In order to tailor the temperature and pressure level needed for a reversible storage of hydrogen to suitable conditions, it is important to understand the mechanism of formation/decomposition of these compounds. An accurate experimental investigation of the electronic charge density distribution of the complex hydrides give important information about the charge transfer and bonding nature between the different atoms within the compounds. Therefore this is a fundamental information for understanding the formation/decomposition mechanisms.

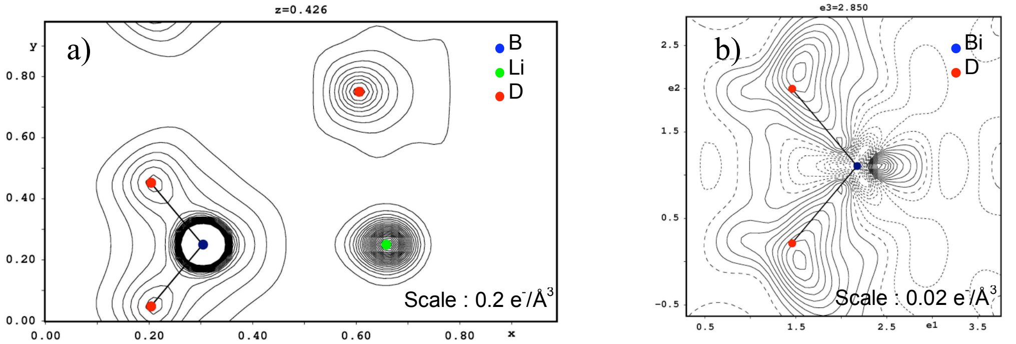

Figure 1. a) Experimental charge density distribution of $LiBD_4$ from maximum entropy method.
b) Difference plot for the charge transfer study within a BD_4 tetrahedron for $LiBD_4$.

Combining neutron diffraction data for the refinement of the structural model, and subsequently X-ray diffraction data for the structure factor extraction, allows the calculation of the experimental charge density distribution[1] (see Figure 1). Due to the instrinsic limited number of extracted structure factors, the maximum entropy method has been used in order to give the most probable charge density map for the set of extracted structure factors[2,3]. The experimental electronic charge density distribution is compared with the electronic charge density distribution obtained for the same compounds by ab-initio calculations[1].

References

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