Neutron Compton scattering from LiH and LiD

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Neutron Compton scattering experiments on various hydrogen containing materials have revealed a striking anomalous deficit in the intensity of the hydrogen recoil peak [1]. The shortfall in intensity ranges from 20% up to 50%, depending on the particular hydrogenous compound, and in most cases shows a dependence on the scattering angle. Initially these experiments were received with great skepticism and questions were raised regarding the correctness of the results and the data analysis. Later detailed control experiments [2] have verified the results and refuted the criticism. Recently, supporting evidence for the effect has been found in electron Compton scattering experiments [3]. Still, this puzzling phenomenon triggers vivid discussions regarding its physical origin, with several different explanations given to date.

In the original publication of Chatzidimitriou-Dreismann et al. on H_2O/D_2O mixtures [4], where such anomalies were reported for the first time, the effect has been attributed to quantum entanglement of the proton with degrees of freedom in its environment. According to these authors, the anomalies in the scattering intensity are due to the extremely short time scale of the collision process (of the order of fs), which is comparable to the lifetime of the entangled proton. Ideas of quantum coherence/decoherence have been pursuit further by Karlsson & Lovesey [5] and Karlsson[6], who presented also rough quantitative estimates of the expected scattering intensity reduction. In a different line of reasoning due to Gidopoulos [7] and Reiter & Platzman [8], the rapid movement of the recoiling proton results in electronic excitations via the breakdown of the Born-Oppenheimer approximation. Thus, scattering intensity is transferred from the recoil peak to higher energies and this leads to the apparent reduction of the recoil intensity.

The motivation for the current work was to perform a detailed and careful study of the phenomenon, on well characterized samples, that would allow us to test specific aspects of the existing theories. For this purpose, LiH and LiD were chosen as sample materials, which are the most studied among the light alkali hydrides and deuterides. The wealth of information currently available on the electronic, lattice, optical and defect properties of these materials will greatly facilitate theoretical model calculations and experimental comparisons. Both lithium hydride and deuteride may be currently obtained commercially in high purity due their technological importance as hydrogen storage materials. It is noted that the substitution of hydrogen with deuterium plays an important role in most of the proposed theoretical explanations and thus represents a very useful tool for this study. From the experimental point of view, lithium hydride and deuteride offer a significant advantage: the light lithium atom has sufficiently high recoil energy so that its scattering signal is clearly discernible from stray signals of heavier atoms (e.g. from sample container, cryostat). This reduces significantly the experimental aberrations.

Neutron scattering measurements were performed at the VESUVIO spectrometer of the ISIS pulsed neutron source. The samples were polycrystalline LiH and LiD powders contained in a thin aluminum sample holder. In a NCS experiment (also called deep inelastic neutron scattering experiment), neutrons with energy in the range of 10–150 eV are scattered by the nuclei in the sample. For scattering on hydrogen and deuterium the momentum and energy transfers are large and the protons (deuterons) are recoiled out of their positions in the crystal where they are situated. In the VESUVIO instrument outgoing neutrons are selected within a narrow energy interval, $E_f = 4.91 \pm 0.14$ eV by the use of Au-197 resonance foils. These neutrons have traveled from the pulsed spallation target at ISIS during a time-of-flight (TOF)

$$t = L_0 / v_0 + L_1 / v_1 ,$$

where v_0 is the velocity of the incoming neutrons and v_1 the velocity of the outgoing ones. The latter is fixed by the resonance foil energy. L_0 and L_1 are the corresponding primary and secondary flight paths, respectively. The TOF at which the recoil peak of a specific nucleus of mass M appears in the spectrum depends on the ratio v_1 / v_0. From classical kinematics it is easy to show that at a scattering angle θ the following expression holds:

$$\frac{v_1}{v_0} = \frac{\cos\theta + \sqrt{(M/m)^2 - \sin^2\theta}}{M/m + 1}$$

where m is the mass of the neutron mass. Fig. 1 shows an example of a NCS spectra taken for LiD at one particular scattering angle, θ = 135°. The recoil peaks from the three different masses are seen: Li, D and Al from the sample container.

Currently the available data have been processed according to the standard procedures of the VESUVIO instrument, which are based on the impulse approximation [9]. The area under the recoil peak for mass *M* is proportional to $N_M \sigma_b$, i.e., the number of atoms times the bound scattering cross-section for the corresponding nucleus. Thus the ratio of the area under the hydrogen or deuterium peak to the area of the Li peak should equal the ratio of the cross-sections $\sigma_{H,D} / \sigma_{Li}$. The nominal values of the cross-section ratios can be easily calculated from cross-section tables and is equal to 59.9 and 5.58 for hydrogen and deuterium, respectively. In the current preliminary phase of data reduction for this very recent experiment, the results obtained for the cross-section ratios are shown in Fig. 2, normalized to their corresponding nominal values. As seen from the figure the normalized values for the cross-section ratios are generally close to unity, indicating that there is no marked intensity anomaly in the LiH/LiD system. This is a striking result in view of previous work on other metal hydrides and deuterides, which all showed an intensity deficit of the proton recoil peak. Further processing of the experimental data is currently under way to assess possible implications of absorption corrections (due to the very large neutron absorption cross-section of Li), multiple scattering and other effects that may affect the cross-section ratio determination.

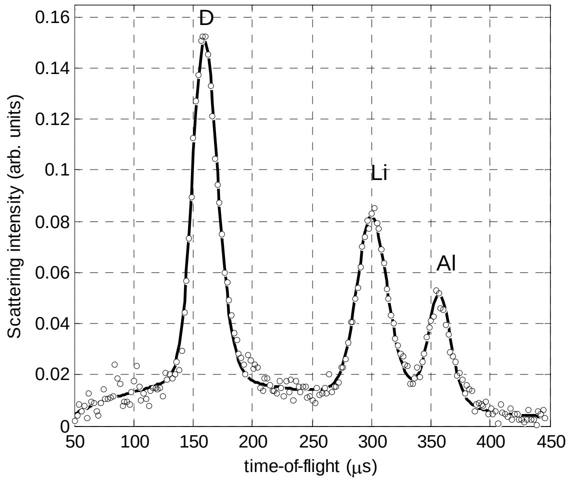

Fig. 1. Typical NCS spectrum. The sample consists of polycrystalline LiD powder in an aluminum container. Circles represent the measured intensity at a scattering angle of 135° as a function of the neutron time-of-flight. The solid line is a fit to the data based on the impulse approximation.

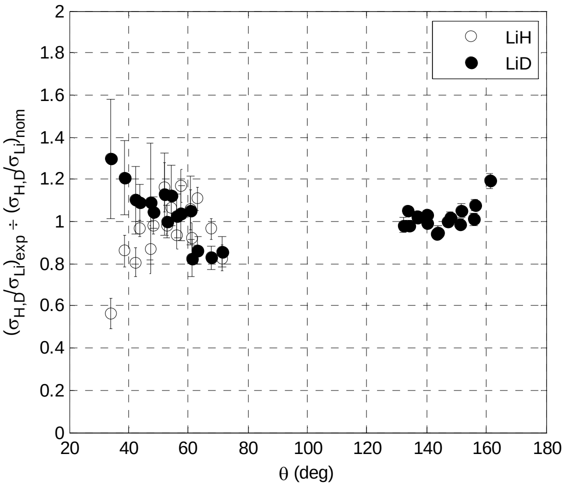

Fig. 2. Experimentally determined ratios $(\sigma_{H,D} / \sigma_{Li})_{\text{exp}}$ normalized to the tabulated ratio $(\sigma_{H,D} / \sigma_{Li})_{nom}$ as a function of scattering angle.

[1] for a review see e.g. C. Andreani et al., *Advances in Physics* **54** (2005) 377.

[2] J. Mayers and T. Abdul-Redah, *Journal of Physics Condensed Matter* **16** (2004) 4811; O. Hartmann et al., *ibid*., **20** (2008) 104248.

[3] C.A. Chatzidimitriou-Dreismann et al., *Phys. Rev. Lett.* **91** (2003) 574031; G. Cooper et al., *ibid*. **100** (2008) 43204.

[4] C.A. Chatzidimitriou-Dreismann et al., *Phys. Rev. Lett.* **79** (1997) 2839.

[5] E.B. Karlsson and S.W. Lovesey, *Phys. Rev. A* **61** (2000) 62714.

[6] E.B. Karlsson, *Phys. Rev. Lett.* **90** (2003) 095301; *Physica Scripta* **77** (2008) 065301.

[7] N.I. Gidopoulos, *Physical Review B* **71** (2005) 054106.

[8] G.F. Reiter and P.M. Platzman, *Physical Review B* **71** (2005) 054107.

[9] J. Mayers and T. Abdul-Redah, *J. Phys.: Condens. Matter* **16** (2004) 4811.

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