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Abstract

Proton Momentum Distributions in Strong Hydrogen Bonds in the Solid State †

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Neutron Compton scattering (NCS) is a unique experimental technique made possible by the development of epithermal neutron sources, such as the ISIS source of the Rutherford Appleton Laboratory in the UK [1,2]. Dynamic structure factors, measured in NCS, are solely determined by the nuclear momentum distribution (NMD). In the picture of purely classical nuclei, the NMD shape is determined by whole energy spectrum of the motional modes, including translational and rotational modes, followed by lattice and internal molecular vibrations. However, more and more experimental evidence has been accumulated over the years that nuclear quantum effects, such as nuclear zero point motion, delocalisation and tunnelling, determine the shapes of NMDs of lightweight isotopes such as protons and deuterons. At sufficiently low temperatures, all nuclear quantum systems are cooled down to their ground states. In this low-temperature limit, the NCS recoil peak shape for a given nucleus is proportional to the square of the absolute value of its nuclear wave function, which is dictated by the shape of the local, effective Born-Oppenheimer (BO) potential [1,2]. Furthermore, different shapes of the BO potentials can be selected by applying Bayesian approach to fitting data obtained from an NCS experiment [3]. Such statistical tests can detect traces of self-interference of a nuclear wave function in effective BO potentials, a prerequisite of nuclear quantum tunnelling in condensed matter systems.

Molecular crystals exhibiting strong hydrogen bonds seem as natural fit for the NCS technique. In this contribution, the results of recent NCS investigation of the solid solutions of equimolar water-phosphoric acid mixture and its deuterated counterpart, will be presented. The analysis of the NMDs, augmented with Bayesian inference methodology, reveals line-shape features characteristic for proton tunnelling in the water-H_3PO_4 mixture below 160 K but shows no such features in the case of the deuterated water-D_3PO_4 mixture. Taken together, these observations suggest the existence of the so-called tunnelling effect in the kinetics of the proton transfer below 160 K, most likely involving concerted proton tunnelling along Grotthuss chains. It is the interplay between the amount of the ZPE and the height of the activation barrier for the proton transfer, which in consequence leads to a non-trivial nuclear quantum isotope effect, whereby kinetic rate constants of protons are orders of magnitude higher than those for deuterium. The presented methodology paves the way for a novel experimental screening protocol for the presence of the signatures of nuclear quantum tunnelling in condensed matter systems.

References

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