Thermal neutron scattering cross sections of liquid deuterium and hydrogen. Applications to cold moderator design. Experiments and simulations.

General outline

Cryogenic liquids have always attracted much interest, owing to both fundamental and technical reasons. “Hydrogens” (i.e. H_2 and D_2), in particular, display a quantum behaviour still challenging theoretical and simulation-based descriptions of the dynamic properties and, at the same time, are the most important moderating systems for the production of cold neutrons.

Despite the importance of these fundamental systems, very slow steps forward have been made, still in the 20th and present centuries, towards the detailed knowledge of their neutron scattering law, or, more generally, of the space-time correlation functions (and their Fourier transforms) describing the dynamic behaviour of these quantum liquids. Inelastic neutron scattering experiments in the (crucial) cold-thermal energy range are incredibly few, published in useless arbitrary units, and lacking in accuracy [1-3]; at the same time, first quantum molecular dynamics simulation methods, attempted to access both the single-molecule (i.e., self) and the collective (i.e., self plus distinct) dynamic correlation functions of great relevance in condensed matter physics, such as the velocity autocorrelation function (VACF) and the (self and total) intermediate scattering functions (ISF). These revealed their limitations, due to the restrictions in computing power (e.g. in the case of VACF calculations) or to the even more serious difficulties brought about, in quantum calculations, by the non-linear dependence on particle positions and momenta of relevant operators (e.g. in the case of ISF calculations).

Preliminary work

Both scientific and applicative motivations make the above-described "hydrogens' situation" an as urgent as appealing case to work on, starting with a research programme able to bridge new experimental efforts and computational/simulation activities: neither of the two being sufficient by itself, but together able to allow for a significant step towards a solution. The boost to such a work is given by a few recent results, the first of which is certainly the validation of quantum Centroid Molecular Dynamics (CMD) as an effective method to simulate the VACF of hydrogen and the detailed probing of the degree of accuracy (and range of applicability) of the Gaussian Approximation (GA) in providing the centre-of-mass self ISF of H_2 from the simulated VACF [4,5]. The second result is the very recent demonstration [6] that quantum CMD simulations of the VACF plus the GA can profitably be employed, within a proper quantum treatment of the intramolecular dynamics [7], to very accurately predict (on an absolute scale and without any adjustment of dynamical parameters) the self contribution to the Double Differential Cross Sections (DDCS) of

liquid hydrogen and deuterium. In other words, this means that, at least for the H_2 case, it is nowadays possible to foresee the scattering behaviour in most kinematic conditions occurring at thermal and higher neutron energies, since the incoherent component largely dominates (except in special conditions we discuss below) the neutron signal from this liquid and, therefore, a good knowledge of the self dynamics alone becomes exhaustive.

The above two findings can be considered as fresh starting points of the further research we now propose here. In particular, they assess our present capability to model the self dynamics of the hydrogens very well and in compliance with the quantum nature of these liquids. A completely open problem is, conversely, how to reach (and exploit) the same confidence in all those cases in which the self dynamics is clearly not sufficient neither to describe the few DDCS available data of D_2 and even H_2 [8], nor to deal, more generally, with the effects of the intermolecular interactions and, consequently, with our full ignorance of the features of the distinct part of the correlation functions of these quantum fluids, which, in addition, still escape an easy access also via simulations.

Scientific aims and Experimental programme

Appropriate ***experimental proposals*** on the Brisp, IN5 and IN6 instruments will be conceived and submitted in order to gather a rich set of inelastic data on the hydrogens. A special concern will be devoted to deuterium because of the increased visibility, due to its coherent scattering, of the sought contributions to the global dynamic behaviour, and because of its more promising use in the development of new-generation cold neutron sources. Nonetheless, scientifically speaking, the H_2 case is even more interesting because of the unpredictable but certainly more consistent quantum effects (due to the lower mass compared to deuterium) which have never been characterized and quantified, until present, as concerns the dynamic properties. Despite hydrogen is the "incoherent" fluid par excellence, measurements of the collective modes of H_2 at equilibrium at liquid temperatures (i.e. in para-H_2) are possible in selected conditions. Indeed, at cold neutron energies or whenever the accessed energy transfers are below the threshold of the first rotational transition ($E_{0\rightarrow1}$~ 15 meV plus the recoil energy), the interplay of initial-state probabilities and spin correlations in para-H_2 actually kills out the incoherent signal, leaving the small coherent cross section at the rudder of both collective and single-molecule contributions. As a result, at limited neutron energies and low Q values, the distinct dynamics warrants a primary role in the response from a nominally "incoherent" system as para-H_2 too. The accurate determination of liquid H_2 low-(Q,E) dynamics, through various measurements, remains of course an experimental challenge, given the adverse conditions for measuring the sought-for (small) coherent signal against background (requiring well-thought measurements, optimized set up of the available spectrometers, and confidence in the preparation of the wanted sample in terms of ortho-para concentration), which merits the success we are quite confident in and which would constitute an inestimably rich and uncommon experimental experience for a willing PhD student under the guide of experts. It is at any rate given for granted that experiments and related proposals must be tailored to the typical times and needs of a three years' PhD work, with experiments and data sets in suitable number to make a student not only able to cope with them, but also to learn detailed analysis procedures and DDCS calculation methods for spin-correlated molecular diatomic liquids as H_2 and D_2.

At the same time, among the scientific aspirations of this project, there is the intent to further

explore and develop ***simulation methods*** of the total ISF of a quantum system, and of the centre-of-mass total correlation functions of hydrogens in particular. As regards this aspect, going-on collaborations have already been established with Prof. M. Neumann of the University of Vienna, with several experts of the Istituto dei Sistemi Complessi of the Italian Centro Nazionale delle Ricerche (CNR) in Florence, and with other researchers of the Dipartimento di Fisica e Astronomia of the University of Florence. The scientific need to develop new and reliable simulation methods of the total dynamics in the quantum case, would certainly profit from the proposed PhD work, since accurate, and presently non-existing, inelastic neutron data are crucial for any validation. Conversely, the proposed PhD work is, in our opinion, already well-sized to take advantage, but not charge, of this further interesting aspect and potentiality we are, parallely, working at.

Last but not least, another scientific aim of the proposed PhD programme is the possibility to verify whether fundamental dynamical parameters, such as the damping and frequency of non-hydrodynamic acoustic excitations, follow, even in quantum fluids, the nearly "universal" behaviour observed in various classical molecular and monatomic liquids [10].

Impact for the ILL – collaboration with other teams

The increase of the neutron cross section data accuracy is of strategic importance for the long term vision of the ILL. Indeed, in parallel to the LEU conversion that is expected around 2030, the whole RHF pile will have to be refurbished. Proposing new cold sources design will thus be crucial before the next international convention in 2023 to give our associates a long term vision. Because the accuracy of the current databases is limited, this design could only be done with the new precise nuclear data (IRSN is involved in the project). The study must start now to be achieved in time.

The previously described, thoroughly scientific, aims of the PhD project also find an encouragement from such interests around source design, whose importance has emerged only in the last few years, for instance with high brilliance directional moderator geometries [14]. Indeed, the deep study of the hydrogen liquids dynamics has become a simultaneous and urgent need to face also the present requests of accuracy in neutron DDCS evaluations and Total Cross Section (TCS) libraries given in use for Nuclear Physics reference databases. In this respect, an accurate knowledge of the neutron DDCS of liquid H_2 and D_2, and of other moderating materials, has become an inescapable requirement for appropriate conduction and development of neutron facilities. However, the existing cross section libraries (ENDF/JEFF...) and scattering kernels employed in nuclear data processing codes [11] are far from being appropriate, especially in the quantum case of hydrogen liquids. The latest investments of the European Community, aimed at remedying the inadequacy of the available cross-section databases, were those allowing the mentioned recent advances and the setting up of the powerful codes in our possess, now able to correctly predict the single-molecule (self) contribution to the DDCS of H_2 and D_2. As mentioned, there are however clear evidences that distinct intermolecular contributions influence the cold neutron signal from para-H_2 [8] and, due to the larger coherent-to-incoherent ratio, this happens even more extensively in the D_2 case, where the neutron response reflects the distinct component also in the thermal range and above. Therefore, a good knowledge of the coherent dynamics of these liquids is an essential ingredient to achieve the accuracy demanded nowadays on neutron cross sections, which is among the objectives of devoted international projects presently in course

[13].

This proposal is complementary with the already granted PhD on UCN production with D_2, which focuses on the very low energy range, whereas we aim to provide thermal neutron scattering cross section information from μeV up to few 100 meV range by combining experiments and simulations.

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- *Support from OECD/NEA for travel and collaboration, in the frame of NAUSICAA [13] ; support from IAEA through the CRP "cold moderator design".*
- *Friendly collaboration with the Munich/ILL current PhD "Low Energy Cross Sections of Liquid and Solid Deuterium" to share knowledge on technical aspects.*
- *Technical support and equipment from the Florence University group.*
- *Collaboration with IRSN for the generation of ENDF/ACE data files and a complementary work on water in the frame of NAUSICAA [13].*

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[13] This proposal is encouraged by the Nausicaa consortium (https://www.ill.eu/?id=15488), gathering the ILL, IRSN, CEA, TUM, ESS, the University of Florence and the Ecole Polytechnique de Montréal, as well as ANL, AECL, IAEA, NEA/OECD and other observers. The core of the Nausicaa project is to measure or properly calculate the double differential cross section of crucial neutron moderators (including water and cryogenic liquids) to produce largely improved S(α,β)-kernels finally available to the nuclear-reactor industry.
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