Critique on the measurement of neutron cross-sections by the Deep Inelastic Neutron Scattering technique

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Abstract. We analyze a recent work of Mayers and Abdul-Redah, [J.Phys.: Condens. Matter 16, 4811 (2004)] in which the authors report the existence of anomalous neutron cross sections in several systems. In the present work we show that the Deep Inelastic Neutron Scattering (DINS) results presented by the authors are affected by an inaccurate formalism employed for obtaining nuclear momentum distributions, and therefore definitive conclusions cannot be drawn on the subject of anomalous cross sections. We also show the reasons why the exact formalism for obtaining momentum distributions that we recently published must be employed for analysing the experimental data instead of the approximations performed in the mentioned work. We also point out serious inconsistencies between different results reported in the mentioned work, as well as incompatibilities with previous results published by the authors. These inconsistencies, as well as experimental evidence against the existence of anomalous cross sections not considered by the authors, reinforce the need to revise critically the procedures on which the usual DINS data analysis is based as well as the proper characterisation of the experimental set-up.

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In a recent work J. Mayers and T. Abdul-Redah presented experimental results obtained with the Deep Inelastic Neutron Scattering (DINS) technique as evidence of the existence of anomalous neutron cross sections in several systems. However, as we have shown in previous papers, the formalism employed by the authors to analyse the experimental data is based on ill-founded approximations, which leads to inaccurate results. This fact created a controversial situation on the real existence of the mentioned anomalous cross sections, which is treated in detail in references. In this comment we will analyse the validity of the conclusions drawn in Ref. , where the authors minimize the importance of errors originated in the approximations assumed in the data analysis. As we will show in this comment, in the authors have omitted important results and conclusions presented in the mentioned previous works, and they have not considered other published works where the addressed issue is analysed. In the following paragraphs, we will itemise the principal reasons wherefore the results and conclusions presented in are flawed, as well as the most important omissions we found in the mentioned work.

(i) Ref. is an important improvement in order to reach a general agreement on the way that DINS experimental data must be treated. It is worth remembering that in previous works the authors have employed the convolution formalism without mentioning its approximated nature. In contrast, in reference the authors admit that the usually employed DINS data treatment (based in a convolution expression) consists in an approximation (CA), that leads to inexact results. However, as a first attempt to analyse DINS experimental data in the proper way, in Ref. the authors present Eq. (2.8) as an alternative form of the exact formalism, which was previously deduced and analysed in , where we have shown that a wide distribution of final energies is operative at every time-of-flight in a DINS spectrum, instead of a single well-defined one as it is assumed in Eq. (2.8). Such analysis clearly shows that the basic hypothesis of a fixed final energy, on which the Eq. (2.8) is based, is wrong.

(ii) In a recently published work we introduced the exact formalism that must be employed for obtaining momentum distributions by DINS to avoid the inaccuracies provoked by the CA. One of the most important results in that work is the demonstration that the exact integration kernel strongly depends on the time of flight. This result shows that the time-of-flight-independent resolution function, usually employed as integration kernel in the CA framework, is inaccurate. As a consequence, the central expression (2.23) of Ref. (also presented by the authors as exact) is not correct and should be replaced by an expression where the probability explicitly depends on time-of-flight $t$.

(iii) It is important to note one of the main results presented in Ref. , namely that the peak intensities obtained with the CA are strongly dependent on the momentum distributions $J(y)$ employed in the fitting process. In Fig. 7 the authors show that CA can introduce a systematic reduction in the obtained ratio $\sigma_H/\sigma_D$. In this
process Gaussian functions for the momentum distributions were assumed. On the other hand, in Fig. 14, we see quite different results when employing non-Gaussian distributions. The results appear to be strongly dependent on the momentum distribution assumed in the fitting process. It is very important to note that these non-Gaussian $J(y)$ were also obtained by the authors in the CA framework [14], and therefore are affected by the already mentioned approximations. In consequence, the results presented in Ref. [1] for $\sigma_H/\sigma_D$ are also affected by CA and a definitive conclusion on the supposed anomalies of the Hydrogen and Deuterium cross sections cannot be drawn.

(iv) It would be most enlightening if the authors presented the results for the Hydrogen, Deuterium, and Oxygen momentum distributions they obtained for the different mixtures (which they did not in Ref. [1]), and also if they compared such results with those previously published. This information should be readily available since it is the primary output of DINS. It is also opportune to remember that important dynamical features such as the mean kinetic energy obtained by DINS, can be independently checked by transmission experiments. As an example, in Ref. [4], we obtained Hydrogen, Deuterium, and Oxygen mean kinetic energies on $H_2O$ and $D_2O$ in excellent agreement with the well-known reference values presented in Refs. [15, 16]. Furthermore, the results obtained for these magnitudes on $H_2O/D_2O$ mixtures are in full agreement with those obtained from linear combination of the different molecular species present in such systems ($H_2O$, $D_2O$ and HDO). Since the momentum distributions assumed in the DINS fitting process have a great influence on the obtained peak intensities, in the future, more efforts should be directed to improve the determination of such distributions. For this purpose, the exact formalism recently presented in Ref. [8] should be the most adequate tool.

(v) The total cross sections of the employed filters are neither Lorentzian nor Gaussian as the authors pointed out. Around each resonance, in the simplest approximation, these are given by the Lamb equation [17]. For a more accurate description, the authors could employ the neutron cross sections available in [18], which were experimentally obtained in a wide energy range.

(vi) The authors omit to mention that the resolution function $R(y)$ they employ is not compatible with the definition of resolution function. The resolution function they employ does not reproduce the neutron Compton profile in the case of an ideal gas at $T= 0$ K. In [2] we have shown that the only resolution function mathematically compatible with the definition of resolution can be analytically deduced from the instrument characteristics. Anyway, we have shown that this resolution function also leads to wrong results, which shows that the problem does not reside in the employed resolution function, but in the convolution formalism itself.

‡ A more detailed description should considered a more realistic nuclear momentum distribution in the filter, where the gas model assumed in the Lamb equation should be replaced by a more detailed model for the solid state dynamics.
(vii) In Ref. [1] the authors mentioned that the reported anomalous cross-sections exhibit different angular behaviours on different systems. For example, in H$_2$O/D$_2$O mixtures the reported anomaly is almost independent of the scattering angle, while in other systems it strongly depends on the scattering angle. In this context, it is important to note the different and inconsistent ways to process the data reported in the literature. More specifically, in the CA framework, the factor $v_1/v_0$ was apparently employed in two different ways. On one hand, Refs. [9, 10] mention a calculation of this factor based on kinematic conditions (i.e. $v_1$ is assumed fixed by the filter and $v_0$ is calculated for each time-of-flight independently of the scattering angle); on the other hand Eq. (3.1) of Ref. [1] (see also Eq. (1) of Ref. [19]) mentions that this factor was calculated according to dynamic conditions (which only depends on the scattered mass and the scattering angle, and are independent of the analysed time-of-flight and the characteristics of the filter). Such discrepancy in the employed data treatments, not mentioned by the authors, could affect the different angular behaviours above mentioned.

(viii) In Fig. 10 the results clearly show a gross systematic difference between the cross section ratio obtained with the single difference method and the one obtained with the double difference method (being the first one about 30% systematically greater than the second one). The authors do not explain the origin of these differences. If the CA employed by the authors were exact the mentioned discrepancies should not exist, since such ratio of neutron cross sections is a constant physical magnitude. Such discrepancy casts serious doubts not only on the formalism employed by the authors for analysing the experimental data, but also on the characterisation performed on the experimental set-up. Furthermore, in Fig. 10 the authors attempt to show that the overlap effect between different peaks is not relevant. However they omit to mention that in [3] we have shown that the inaccuracies of CA for obtaining peak intensities are still present even when the intensity of an isolated peak is analysed, i.e. when the overlap effect is absent.

(ix) Some results presented by the authors are incompatible with those published in previous works. For example, in reference [13] it is concluded that the reduction reported for the ratio $\sigma_H/\sigma_D$ in H$_2$O/D$_2$O mixtures is only originated in a reduction of $\sigma_H$, while the obtained value of $\sigma_D$ agrees with the tabulated value. On the other hand, in Fig. 6 opposite results are presented, i.e. an anomaly in the Deuterium intensity and not in Hydrogen.

The validity of the DINS results presented in [1] not only depends on the formalism employed for analysing the experimental data, but also on a proper characterisation of the experimental set-up. In [1] the authors have performed a characterisation of different components of the experimental set-up, which in the light of the inconsistencies observed, could be improved, verified, and/or performed by alternative methods never employed in VESUVIO. Details of such alternative characterisation, as well as additional disagreements between different results presented in [1], will be discussed elsewhere.
We wish to remember that, in order to investigate the cross sections of Hydrogen and Deuterium in light water/heavy water mixtures, we performed transmission experiments on such mixtures on the epithermal neutron energy range employing the Bariloche Electron LINAC \[4\]. Our experiment shows no traces of anomalous neutron cross sections, and the values we obtained are in perfect agreement with the tabulated data. As was explained in that work, our transmission results are conclusive evidence on the absence of anomalous neutron cross sections in the mentioned mixtures. It must be noted that both techniques, transmission and DINS, measure the same magnitude in exactly the same sample and scattering conditions. Due to the reason exposed in \[4\], any anomaly in the bound-atom cross section observed by DINS should also be observed in transmission experiments. The authors of Ref. \[1\] neither mention that results, nor the arguments on this subject presented in Ref. \[20\], where we have shown that the assertions of Ref. \[21\] are wrong.

Finally, it is worth to mention that the absence of anomalous neutron cross sections in H\(_2\)O/D\(_2\)O mixtures was very recently confirmed by scattering experiments carried out by an independent group \[22\]. These results agree with those we obtained by transmission \[4\], as well as with those obtained by precise interferometric techniques \[23\]. In summary, in order to analyse the anomalies reported/suggested in \[12\], three independent techniques were applied by three independent groups, and always-negative results were obtained.

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