Journal of Alloys and Compounds 404–406 (2005) 790–793

Journal of ALLOYS AND COMPOUNDS

www.elsevier.com/locate/jallcom

Anomalous neutron Compton scattering cross section in zirconium hydride

T. Abdul-Redah[a,b], M. Krzystyniak[c,*], J. Mayers[b], C.A. Chatzidimitriou-Dreismann[c]

[a] *School of Physical Sciences, University of Kent at Canterbury, Canterbury, UK*
[b] *ISIS Facility, Rutherford Appleton Laboratory, Chilton/Didcot, OX11 0QX, UK*
[c] *Institute of Chemistry, Stranski Lab. ER1, TU Berlin, D-10623 Berlin, Germany*

Received 7 June 2004
Available online 15 July 2005

Abstract

In the last few years we observed a shortfall of intensity of neutrons scattered from protons in various materials including metal hydrogen systems using neutron Compton scattering (NCS) on the VESUVIO instrument (ISIS, UK). This anomaly has been attributed to the existence of short-lived quantum entangled states of protons in these materials. Here we report on results of very recent NCS measurements on ZrH_2 at room temperature. Also here an anomalous shortfall of scattering intensity due to protons is observed. In contrast to previous experiments on $NbH_{0.8}$, the anomalies found in ZrH_2 are independent of the scattering angle (or momentum transfer). These different results are discussed in the light of recent criticisms and experimental tests related to the data analysis procedure on VESUVIO.

Keywords: Hydrogen storage materials; Neutron Compton scattering; Attosecond quantum effects and decoherence

1. Introduction

In recent years, many neutron Compton scattering (NCS) experiments on various hydrogen containing materials like water [1], organic compounds [2–5] and also metal hydrogen systems [6–9] have been reported. A significant deficit of the scattering intensity due to the protons has been found in these experiments. For example in experiments on H_2O/D_2O mixtures [1], the peak area ratio of H to that of D does not scale with the number density of particles in the sample. Rather, the area ratio normalized to the number density, i.e. σ_H/σ_D, was significantly smaller than the expected ratio of 10.7 and strongly dependent on the D mole fraction in the mixture. Several theoretical models have been suggested to explain this striking effect. They propose the existence of short-lived quantum entanglement of protons in condensed matter [10–12] and/or refer to the breakdown of the Born–Oppenheimer approximation during the scattering process [12–14].

The measurements were performed using the time of flight inverse geometry spectrometer VESUVIO (see Fig. 1A) at ISIS/UK. The polychromatic epithermal neutrons are scattered by the sample into detectors arranged in an angular range $30^\circ < \Theta < 80^\circ$. An analyzer foil situated between sample and detectors is cycled in and out of the sample detector axis. The analyzer foil absorbs neutrons at a fixed energy, e.g. 4.9 eV if using gold. The difference of a foil-in and a foil-out spectrum gives the final spectrum to be analysed. It is important to note that the final energy E_1 is fixed while the incident energy E_0 is variable and is governed by the kinematic conditions as well as by the masses of the scattering nuclei. Due to the high transfers of momentum $q = |\mathbf{k}_0 - \mathbf{k}_1|$ (where $\mathbf{k}_0$ and $\mathbf{k}_1$ are the incident and scattered neutron wave vectors, respectively) and energy $\omega = E_0 - E_1$ the scattering of the neutrons by the nuclei is impulsive. As a consequence of this so called impulse approximation (IA) limit, the dynamic structure factor $S(q, \omega)$ of a nucleus of a particular mass M consists of a single peak, i.e., $S(q, \omega) = J_M(y_M)M/\hbar q$. Here y_M is a scaling variable $y_M = (\hbar\omega - \hbar^2 q^2/2M)M(\hbar q)^{-1}$ [15] and

---

[*] Corresponding author. Tel.: +49 30 314 23735; fax: +49 30 314 26602.
E-mail address: krzystyniak@iwan.chem.tu-berlin.de (M. Krzystyniak).

0925-8388/$ – see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.jallcom.2005.05.003

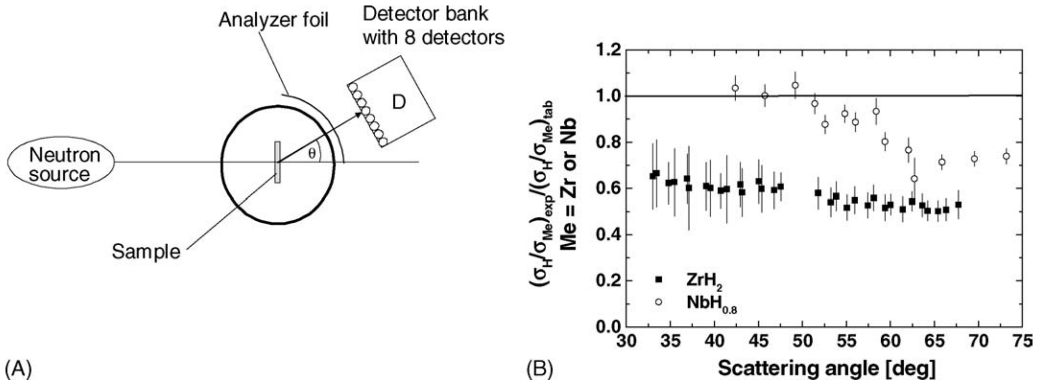

Fig. 1. (A) Schematic instrument setup: the polychromatic neutrons leave the neutron source, are scattered by the sample under the angle θ and are detected in D. The analyzer foil is cycled in and out of the scattered neutron beam. (B) The experimental ratios $(\sigma_H/\sigma_{Zr})_{exp}$ (full squares) and $(\sigma_H/\sigma_{Nb})_{exp}$ (open circles) of the scattering cross section of H to that of Zr and Nb of ZrH_2 and $NbH_{0.8}$, respectively, each one normalized to its tabulated value (i.e. $(\sigma_H/\sigma_{Zr})_{tab}$ and $(\sigma_H/\sigma_{Nb})_{tab}$) as a function of scattering angle θ. Whereas the ZrH_2 data are very flat, the $NbH_{0.8}$ show strong scattering angle dependence although the same Jacobian (see Fig. 2) and the same incident neutron intensities are involved for both ZrH_2 and $NbH_{0.8}$. The $NbH_{0.8}$ data are adapted from Ref. [6].

$J_M(y_M)$ is the neutron Compton profile of the scattering nucleus which is assumed to be of Gaussian shape. Using these quantities, the double differential scattering cross section is $d^2\sigma/d\Omega\, dE_1 = b_M^2 J_M(y_M) M k_1/(\hbar q k_0)$. The experimentally observed intensity is then proportional to $J_M(y_M)$ convoluted with the instrument resolution function $R_M(y_M)$, i.e. the total number of neutron detected in a time channel is [16]:

$$c(t) = \frac{E_0 I(E_0)}{q} \sum_M B N_M b_M^2 M J_M(y_M) \otimes R_M(y_M) \qquad (1)$$

where B contains instrument parameters and b is the scattering length. Thus, the peak area $A = B N_M b_M^2$ is proportional to the number density N and the scattering cross section σ of a nucleus in the sample. It follows that [16]:

$$\frac{A_i}{A_j} = \frac{N_i b_i^2}{N_j b_j^2} = \frac{N_i \sigma_i}{N_j \sigma_j} \qquad (2)$$

2. Experimental and results

We performed new NCS measurements on ZrH_2 at room temperature on VESUVIO. The sample was put in a standard flat Al can. Using the back scattering detectors – where even higher energy and momentum transfers are involved in the scattering – it is possible to resolve the Zr ($M = 91$) peak from the Al ($M = 27$) one. This facilitates the determination of the ratio σ_H/σ_{Zr} from the forward scattering spectra (for more details, see Refs. [3,4]). The experimental ratios of the scattering cross sections have been determined for all available detectors separately in forward scattering using Eq. (2). The experimental results show that ZrH_2 (see Fig. 1B, full squares) exhibits a strong anomalous shortfall. For the sake of better comparability, depicted are the experimentally determined values divided by the tabulated ones, i.e. $(\sigma_H/\sigma_{Zr})_{exp}/(\sigma_H/\sigma_{Zr})_{tab}$. As one can see very easily, these values differ from unity by a considerable amount. Furthermore, these values are almost independent of the scattering angle (or momentum transfer).

3. Summary and conclusions

The presented experimental result provides a further support of the striking phenomenon of the neutron scattering cross section anomaly for protons in condensed matter. It is very interesting to compare the ZrH_2 results with previous results on the metallic hydride $NbH_{0.8}$ [6]. $NbH_{0.8}$ (see Fig. 1B, open circles) and ZrH_2 show completely different results: (1) while the largest anomaly found in $NbH_{0.8}$ is about 30%, an intensity deficit of ca. 45% is observed for ZrH_2; (2) in contrast to the $NbH_{0.8}$ results, ZrH_2 does not show a significant angle dependence. That is, neither the angle dependence nor the magnitude of the anomaly of $NbH_{0.8}$ is reproduced by the results of ZrH_2.

This comparison is very interesting in the light of recent criticisms. It was suggested by Cowley [17] that the angular dependent anomaly of σ_H/σ_{Nb} of $NbH_{0.8}$ might be caused by the lack of proper data corrections due to the incident neutron flux $I(E_0)$ (see Eq. (1)) or due to a Jacobian factor $J = 1 - (1 - \cos(\Theta) k_1/k_0) m/M$ involved in the conversion of a time of flight spectrum into q space (m: neutron mass). It is very important to note that the masses of Nb and Zr are almost equal, i.e. $M_{Nb} = 93$ and $M_{Zr} = 91$ a.u. This leads to the fact that the same energy transfer is involved in scattering on Zr and Nb. For this reason and because the final energy E_1 is fixed, the same incident neutron energies E_0 are involved for scattering on Zr and Nb (see Fig. 2A). Consequently, the same correction of incident neutron spectrum is involved in the data analysis. In addition, again due to the similarity of the masses of Zr and Nb, the same Jacobians are involved for the scattering on those nuclei (see Fig. 2B). This means that, despite the very similar masses of Zr and Nb, the dependence of the scattering cross section from protons on

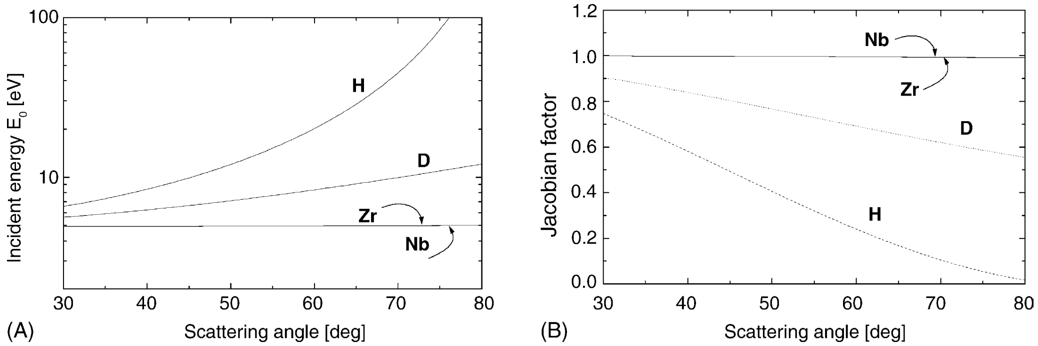

Fig. 2. (A) Dependence of the involved incident energy on the scattering angle θ for different masses: H ($M = 1$), D ($M = 2$), Zr ($M = 91$), and Nb ($M = 93$). The same incident energy is involved for Zr and Nb. (B) Dependence of the involved Jacobians ($J = 1 - (1 - \cos(\Theta)k_1/k_0)m/M$) for H, D, Zr, and Nb. The same Jacobian factor is involved for both, Zr and Nb.

the scattering angle is totally different for ZrH_2 and $NbH_{0.8}$. Thus, we may conclude that the involved different Jacobians do not artificially cause the strong deviations of σ_H/σ_{Zr} from the tabulated value.

Rather, the angle dependence in the case of $NbH_{0.8}$ might be interpreted in the following way. Within the IA, it is possible to define a scattering time by $q(\theta)\tau_{sc}v_0 \approx 1$ [15]. Here $q(\theta)$ is the scattering angle dependent momentum transfer, and v_0 is the root mean square velocity of the nucleus before scattering. τ_{sc} can be regarded as the time window within which the dynamics of the nucleus is sampled. In the case of $NbH_{0.8}$, the anomalies are large at short scattering times ($<0.5 \times 10^{-15}$ s) and seem to vanish for long τ_{sc}. If these anomalies are attributed to the existence of short-lived quantum entangled particles involving mainly the protons, this τ_{sc} dependence may be interpreted such that at short τ_{sc} quantum interferences are revealed while at longer τ_{sc} a time average is taken thus leading to the disappearance of the anomalies. However, Cowley argued further that it is essential that the energy resolution is better than the energy splitting of two entangled spin configurations [17]. As the energy resolution of the VESUVIO spectrometer is about 0.1 eV i.e. much larger than any quantum splitting of the energy levels in solids, it was claimed that this spectrometer cannot reveal any effects of quantum entanglement. However, this statement is not always correct: e.g., as is well known, thermal neutrons scattered from, or transmitted through, liquid H_2 are able to distinguish between *ortho*- and *para*-H_2, because of the strongly different total cross sections (cf. e.g. Ref. [18]) independently of any resolution requirements.

In addition, it was stressed [17] that the existing theories [10–12] explaining σ_H decrease in experiments with VESUVIO should be incorrect because they are inconsistent with the first-moment sum rule (FMSR) for $S(q, \omega)$ [18]. On the one side, there are different theoretical models [13,14] which are consistent with the FMSR. On the other side, as pointed out in Ref. [12], the scattering systems under consideration ought to be described as open quantum systems and consequently they always exhibit decoherence and thus a non-unitary time evolution. However, the derivations of the sum rules are based on unitary quantum dynamics (which do describe closed systems; see textbooks, e.g., Ref. [18]).

In summary, the present results reveal a strong anomalous shortfall of intensity of neutrons scattered by protons in ZrH_2 at room temperature. The scattering angle independent anomaly together with the strong angle dependent anomaly previously found in $NbH_{0.8}$ indicate further that possible inaccuracies of the data reduction procedure (e.g., due to the incident neutron beam spectrum and involved Jacobians) cannot account for the found anomalies. The anomalies revealed in various hydrogen containing materials have found thus far no consistent explanation within existing condensed matter theories. Very recent experimental results e.g. on LiH [9] suggest that the electronic environment surrounding the hydrogen atom in the material could be responsible for the different features of the cross section anomalies of the proton. More systematic experiments on materials with adjustable electronic structures are necessary in order to shed more light onto their influence on the decoherence process of the protons. Let us also mention that, very recently, this NCS effect was confirmed by applying an independent method, namely electron–proton Compton scattering (ECS) [5]. This method is considerably different from NCS due to the fact that the Coulomb interaction is involved in contrast to the strong interaction involved in NCS [5]. In addition, the apparatus with which the ECS experiments have been performed is completely different in that the incoming electron beam is monochromatic and the spectra are recorded with respect to energy loss.

References

[1] C.A. Chatzidimitriou-Dreismann, T. Abdul-Redah, R.M.F. Streffer, J. Mayers, Phys. Rev. Lett. 79 (1997) 2839.
[2] C.A. Chatzidimitriou-Dreismann, T. Abdul-Redah, J. Sperling, J. Chem. Phys. 113 (2000) 2784.
[3] C.A. Chatzidimitriou-Dreismann, T. Abdul-Redah, B. Kolaric, J. Am. Chem. Soc. 123 (2001) 11945.
[4] C.A. Chatzidimitriou-Dreismann, T. Abdul Redah, R.M.F. Streffer, J. Mayers, J. Chem. Phys. 116 (2002) 1511.

[5] (a) C.A. Chatzidimitriou-Dreismann, M. Vos, C. Kleiner, T. Abdul Redah, Phys. Rev. Lett. 91 (2003) 057403;
(b) C.A. Chatzidimitriou-Dreismann, M. Vos, C. Kleiner, T. Abdul Redah, Phys. Today Phys. Update (2003) 9;
(c) C.A. Chatzidimitriou-Dreismann, M. Vos, C. Kleiner, T. Abdul Redah, Scientific Am. (2003) 34.

[6] E.B. Karlsson, C.A. Chatzidimitriou-Dreismann, T. Abdul-Redah, R.M.F. Streffer, B. Hjörvarsson, J. Öhrmalm, J. Mayers, Europhys. Lett. 46 (1999) 617.

[7] T. Abdul-Redah, R.M.F. Streffer, C.A. Chatzidimitriou-Dreismann, B. Hjörvarsson, E.B. Karlsson, J. Mayers, Physica B 276–278 (2000) 824.

[8] E.B. Karlsson, T. Abdul-Redah, R.M. Streffer, B. Hjörvarsson, J. Mayers, C.A. Chatzidimitriou-Dreismann, Phys. Rev. B 67 (2003) 184108.

[9] T. Abdul-Redah, C.A. Chatzidimitriou-Dreismann, Physica B 350 (2004) e1035.

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

[11] E.B. Karlsson, S.W. Lovesey, Physica Scripta 65 (2002) 112.

[12] C.A. Chatzidimitriou-Dreismann, J. Alloys Comp. 356–357 (2003) 244.

[13] G.F. Reiter, P.M. Platzman, Phys. Rev. B 71 (2005) 54107.

[14] N. Gidopoulos, Phys. Rev. B 71 (2005) 54106.

[15] V.F. Sears, Phys. Rev. B 30 (1984) 44.

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

[17] R.A. Cowley, J. Phys.: Condens. Matter 15 (2003) 4143.

[18] G.L. Squires, Introduction to the Theory of Thermal Neutron Scattering, Dover Publications, Mineola, NY, 1978.