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LOW-FREQUENCY EXCITATIONS IN ZIRCONIUM HYDRIDES

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INTRODUCTION

Hydrogen in zirconium has been extensively studied over the last few decades both from the fundamental and applied physics point of view [1].

The theoretical studies and the dynamics of hydrogen (deuterium) in Zr investigated especially by neutron scattering techniques allowed to obtain information concerning: (i) the diffusional motions, by examining the width of the quasielastic incoherent scattering in the range of very low energy transfers [2], (ii) the vibrations of H (or D) atoms within the range of the host lattice dynamics by analysing the inelastic scattering spectra in the energy range less than 30 meV [3-6] and (iii) the local vibrations of H (or D) by measuring the high energy spectra in the range of optical phonons [6-11].

The interest for dynamical properties has been devoted mainly to the high energy H(D) vibrations which seem to be at the present quite well understood. The low-energy H vibration spectra earlier obtained by INS were not enough analysed, most of the results being related with the acoustic phonons of Zr lattices. The present INS measurements provide new information to compare with theoretical models. A simple theoretical model [12] assumed the influence of some hypotetical low-frequency H(D) local modes of tunneling or resonance type in INS experiments.

Low-frequency modes have been also theoretically predicted in terms of resonant vibrations but for H dissolved in V, Nb, Ta and Pd [13]. Generally speaking, such as vibrations are common for heavy deffects in metallic lattices and their occurrence in the case of interstitial H in metals is still not fully elucidated. So far, H resonant vibrations have been observed in neutron scattering studies of $Pd_{0.9}Ag_{0.1}H_{0.2}$ [14] and $NbH_{0.05}$ [15].

However, neutron scattering experiments on Zr hydrides over a large range of H concentrations for low H contents show an unusual low-energy dynamics. A such as feature could be interpreted as possible H resonant vibrations in Zr lattice [4-5]. In order to investigate this effect with a better resolution a new experimental study has been performed on several ZrH_x systems by means of incoherent inelastic cold neutron scattering at DIN-2PI time-of-flight spectrometer, IBR-2 pulsed reactor of JINR-Dubna.

We report below the measurements and the results of this study obtained in the range of low energies in terms of ($d^2\sigma$ / $d\Omega dE$) the double differential cross-section and $g(\hbar\omega)$, the generalized vibrational density of states (GVDS).

EXPERIMENTS

The inelastic neutron scattering investigations were carried out on ZrH_x systems with x= 0.38 and 0.52 by means of the DIN-2PI time-of-flight spectrometer installed at the IBR-2 pulsed reactor of JINR-Dubna. The samples including both the disordered solid-solution of H in α-HCP Zr and the ordered γ-hydride with a FCO Zr lattice were pollycrystalline powders sealed into thin foil Al flat containers. Measurements were made with an incident neutrons energy of 4.39 meV that allowed to obtain a resolution of 0.28 meV (full width at half maximum of the elastic scattering taken on a vanadium sample). An accurate analysis of dynamical features in the energy range of less 30 meV became possible. The data were collected in a large angular range between 11°-134°. After applying all typical corrections the data were normalysed to the elastic scattering of vanadium and converted to the double differential cross section ($d^2\sigma$ / $d\Omega dE$). The GVDS has been derived according to the monophononic approximation

$$\frac{d^2\sigma}{d\Omega dE} = \frac{\sigma_b}{4\pi} g(\hbar\omega) e^{-2W} \sqrt{\frac{E}{E_0}} \frac{E + E_0 - 2\cos\theta \sqrt{EE_0}}{\hbar\omega\left(exp(\hbar\omega / k_B T) - 1\right)} \quad (1)$$

whith $\hbar\omega = E - E_0$, 2W the Debye-Waller factor, K_B Boltmann's constant.

RESULTS

The double differential cross section in the range of low energies is shown in Figures 1 - 2 at two scattering angles. As it is seen all the spectra exhibit a sharp feature located around 15 meV and a broader one around 25 meV. Other features whose presence becomes weaker at higher H content are also observed around 11 meV and 18 meV. In addition to the expected spectral components located in the range of acoustic vibrations of Zr atoms in various regular lattices of the samples other unexpected features are also revealed at low-energies, between 2-10 meV. For $ZrH_{0.38}$ these features show a good resolved fine structure as peaks at 2.5 meV and 6 meV while for $ZrH_{0.52}$ the presence of this structure is much less evident. This clearly revealed low-energy effect could be related to the observations reported by a worse resolution in previous experiments [4-5]. An analysis of the GVDS spectra in parallel with that of metallic Zr [16] (Figures 3-4) allowed to give an interpretation to the peaks located at energies higher than 10 meV. The latter one are identified as Zr acoustic vibrations both in a HCP lattice (those located at 11 and 18 meV) and in a FCO one (those located at 15 and 25 meV). The new features revealed in $d^2\sigma$ / $d\Omega dE$ spectra at energies lower than 10 meV cannot be assigned to the lattice vibration. However, their dependence of the H content is an evidence of excitations undoubtedly related to H dynamics. A such low energy H dynamics is represented either by tunneling or resonant modes. These are typical for H dissolved in metals as it was theoretically shown for resonances in H-(V, Nb, Ta and Pd) systems [13] and for tunneling effects in H(D)-(Zr, Hf) systems [12]. Unlike the tunneling motions predicted only for H solid solutions in Zr or Hf (values of concentration of the order of 10^{-2} to 10^{-1}), the resonantlike vibrations of H or D were observed by neutron scattering for both H solid solutions in metals - $NbH_{0.05}$ [15] and for high H containing hydrides - $Pd_{0.9}AgH_{0.2}$ [14], $NbD_{0.85}$ and $NbH_{0.82}$ [17].

To bring more light on the origin of the new observed peaks in terms of H tunnelling or resonance effect in α-Zr lattice new investigations are needed.

CONCLUSIONS

We have measured with a good resolution the slow inelastic neutron scattering spectra on ZrH_x at various H concentration. From the analysis of INS in terms of double differential cross section and of GVDS it is found that the low energy transfer spectrum shows new feature not observed in the previous experiments. Thus the excitations detected at energies lower than 10 meV are undoubtedly related to the proton dynamics and could be interpreted in terms of a resonantlike vibration. In order to elucidate these new foundings new experiments are planned at lower temperatures.

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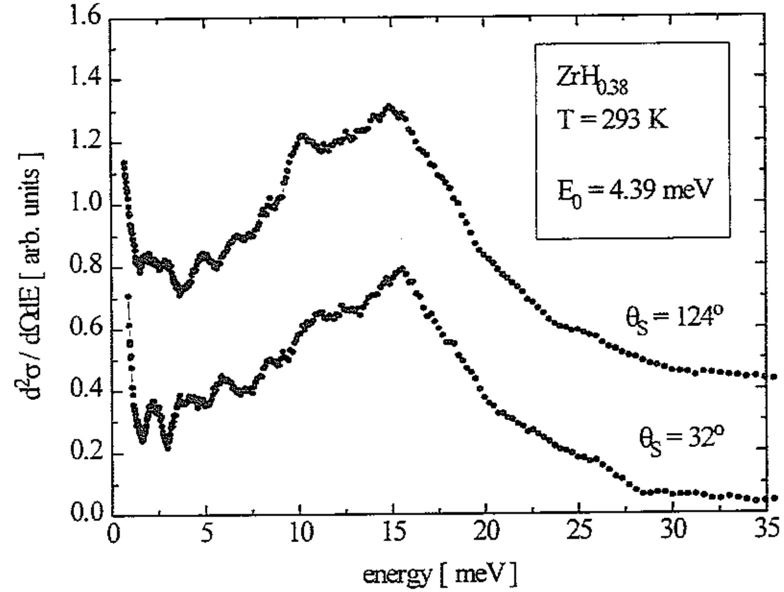

Fig. 1 - The double differential cross sections for $ZrH_{0.38}$;

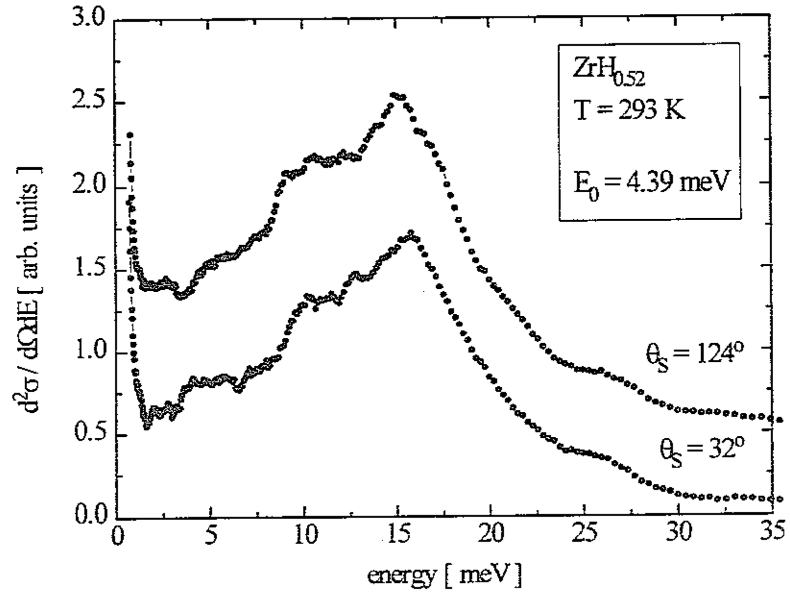

Fig. 2 - The double differential cross sections for $ZrH_{0.52}$;

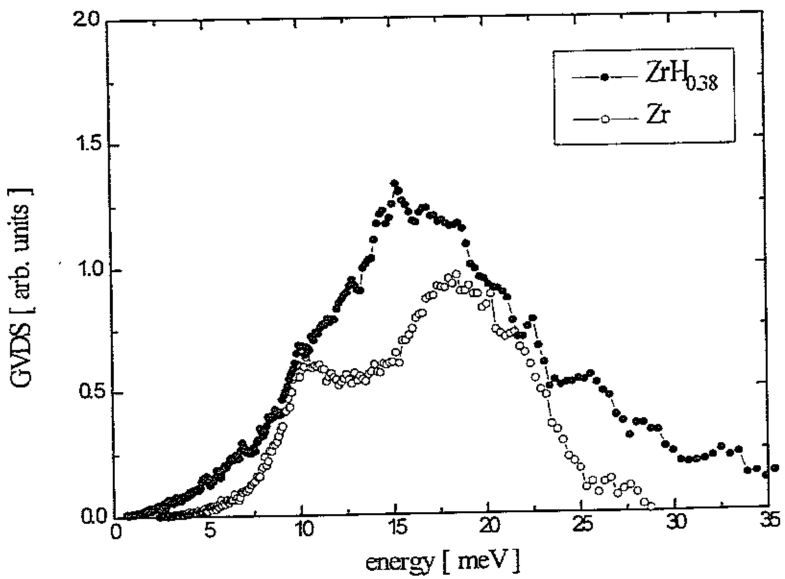

Fig. 3 - GVDS for $ZrH_{0.38}$ compared with that for α-Zr;

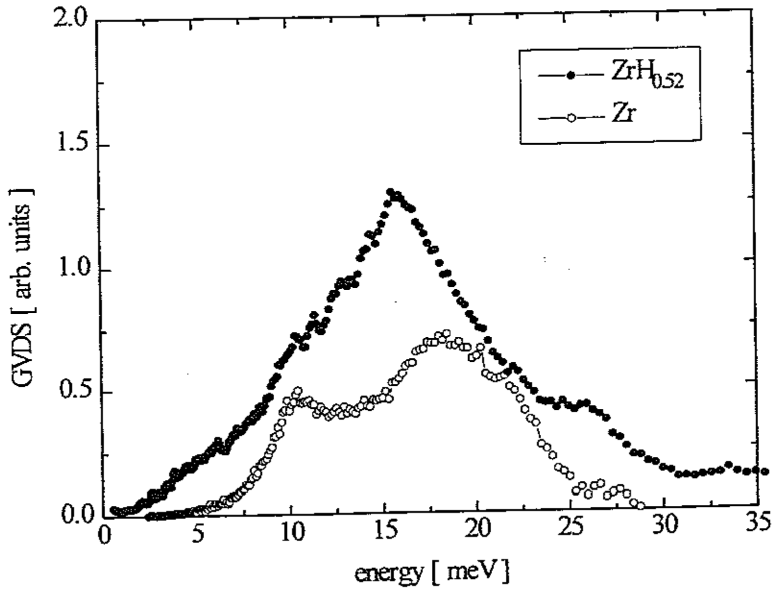

Fig. 4 - GVDS for $ZrH_{0.52}$ compared with that for α-Zr;

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