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Effects of Chemical Binding on the Neutron Cross Section of Hydrogen†

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Fermi derived the energy dependence of the neutron scattering cross section of hydrogen chemically bound such that it acts as a harmonic oscillator. For comparison with this theory, the total cross section was measured with a high-resolution crystal spectrometer for hydrogen bound in four materials. In zirconium and yttrium hydrides, in which chemical bonding is relatively isotropic, the predicted structure with discrete energy levels is observed. In the less isotropic magnesium hydride, there is some evidence of energy levels, although not clearly resolved. For water the variation in the cross section is smooth, with no evidence of energy levels.

THE total cross section decreases with increasing neutron energy E from a value characteristic of the fully bound atom, σ_b, to a value characteristic of the free atom, σ_f. The ratio of these cross sections is given by $\sigma_b/\sigma_f=(A+1)^2/A^2$, where A is the atomic mass of the scattering atom. Fermi[1] treated the manner in which the cross section of hydrogen varies from the free atom value to the fully bound value in the energy region where chemical binding effects are of importance. In his treatment, the hydrogen atom is considered as bound to an infinitely heavy atom and executes isotropic harmonic oscillations with a single sharp frequency ν. This implies a set of energy levels equally spaced by the value $h\nu$. For this case, Fermi showed the form of the expected variation of the cross section with energy to be that shown in Fig. 1.

As the incident energy increases, the cross section σ falls rapidly from the fully bound cross section $(\sigma=4\sigma_f)$ to a sharp minimum value $\sigma=\sigma_f$ at the threshold of the first level. It then varies through a series of peaks of decreasing amplitude, returning to the free hydrogen cross section at the threshold of each level. The amplitude of these peaks continuously decreases with increasing n until finally the cross section is just the free hydrogen cross section for $E\gg h\nu$. Even for an ideal substance the sharp minima will be spread out because of the thermal motion of the atoms. Figure 2 shows the magnitude of the effect on the first minimum for the case of zirconium $(M=91)$ and magnesium $(M=24)$. There is a similar effect for the minima occurring at higher energies.

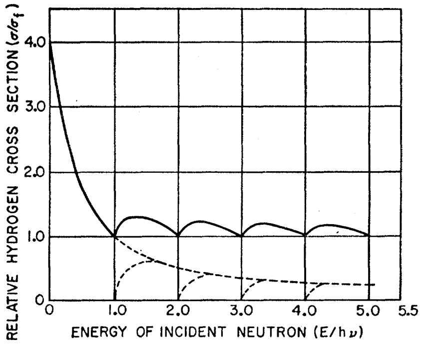

Fig. 1. Theoretical neutron cross section for a bound proton, with frequency ν of the isotropic harmonic oscillation, plotted in terms of the free hydrogen cross section σ_f.

The total hydrogen cross section as a function of the energy of the incident neutron has been measured for zirconium hydride. Since previous measurements[2] indicated that the hydrogen in zirconium hydride executes harmonic oscillations with a rather well-defined frequency, zirconium hydride should exhibit the effects predicted above. Figure 3 shows the total

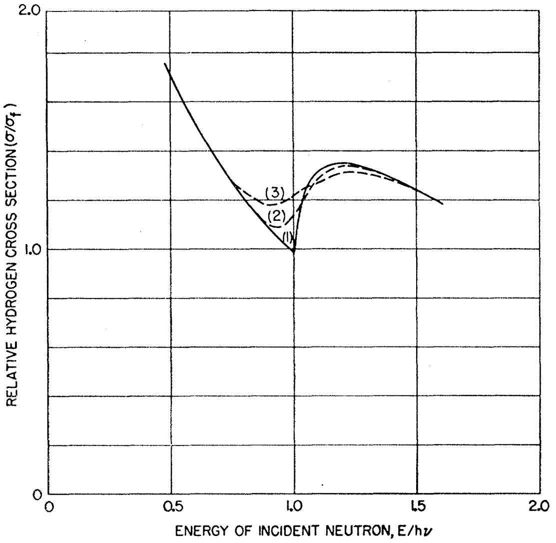

Fig. 2. Theoretical neutron cross section of bound proton, taking account of the thermal motion of the crystal lattice. Cross sections plotted in terms of the free hydrogen cross section, σ_f. Curve 1 for an infinite mass (Fermi's curve); curve 2 for zirconium hydride; curve 3 for magnesium hydride.

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[1] E. Fermi, Ricerca sci. **7**, 13 (1936).

[2] Pelah, Eisenhauer, Hughes, and Palevsky, Phys. Rev. **108**, 1091 (1957); Andresen, McReynolds, Nelkin, Rosenbluth, and Whittemore, Phys. Rev. **108**, 1092 (1957).

cross section per hydrogen atom obtained for a sample of zirconium hydride ($ZrH_{1.5}$) by a high-resolution neutron crystal spectrometer specially constructed to give accurate neutron cross sections. Three well-formed peaks appear in the cross section and are due to the excitation of the first three energy levels of the hydrogen oscillator. The constant cross section for zirconium has been deducted from the measured cross section to give the results shown. The absolute value of the cross section can be in error by 5% because of uncertainty in the concentration of hydrogen in the hydride sample.

In the previous measurements,[2] the value of the harmonic energy level $h\nu$ was determined to be 0.130±0.005 ev. From the theoretical curves shown in Figs. 1 and 2, it is expected that the cross section should have a minimum near this energy and other minima located near to integral multiples of this energy. Such minima do appear in Fig. 3. The theoretical curve from Fig. 2 for zirconium hydride has been fitted to the experimental data by treating $h\nu$ as an arbitrary parameter to be adjusted to give a satisfactory agreement between the experimental and theoretical curves. A value for $h\nu$ of 0.137 ev gives such an agreement. This value agrees within the precision of the experiments with the value of 0.130±0.005 obtained before. It is seen that for the region below the first harmonic level the theoretical and experimental curves agree exactly. The curves deviate somewhat in the region of the first and second maxima. This deviation is worse for the second maximum, as would be expected if the isotropic harmonic oscillator model used for the theoretical curve is not accurate.

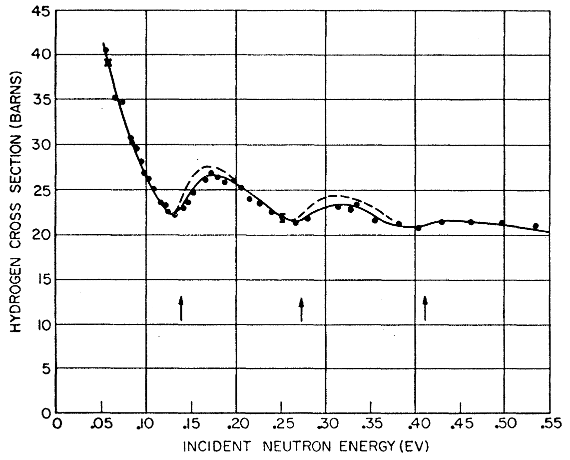

FIG. 3. The solid curve gives the neutron cross section per hydrogen atom in zirconium hydride ($ZrH_{1.5}$). Arrows indicate the excitation levels for H vibrations in Zr lattice. The Fermi theoretical curve corrected for thermal motion has been fitted to the above data with $h\nu = 0.137$ ev and agrees with the experimental curve except as shown by the dashed curve. Experimental errors are indicated.

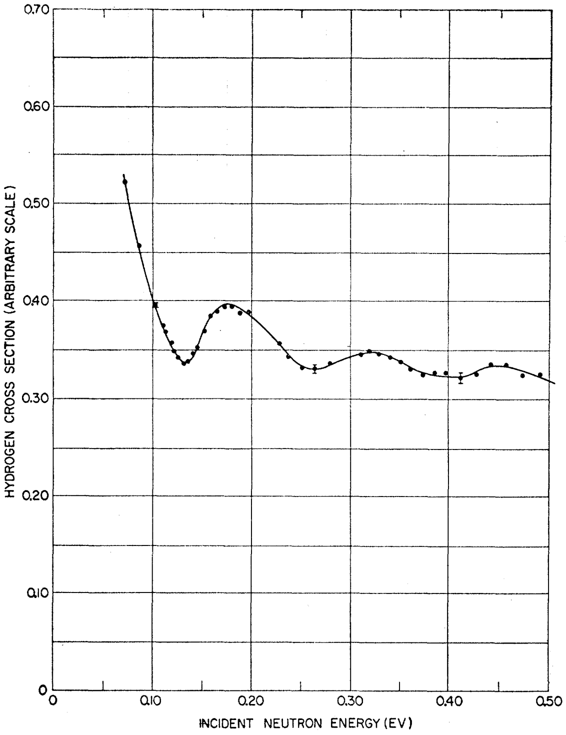

FIG. 4. Total neutron cross section for hydrogen in yttrium hydride, plotted on an arbitrary scale. The well-resolved peaks indicate isotropic hydrogen oscillations of about the same frequency as in zirconium hydride. Experimental errors are shown.

Further measurements of the above type were made for yttrium hydride, magnesium hydride, and water. The cross section for the hydrogen in the metal hydrides should demonstrate the above behavior in varying degrees depending on the extent to which the hydrogen oscillator is isotropic. Figure 4 shows the relative hydrogen cross section for yttrium hydride. Since the concentration of hydrogen in the hydride was uncertain, the cross section per hydrogen atom is not computed. The experimental details of this curve are exactly the same as those for zirconium hydride. Since the cross section exhibits the same well-formed peaks at the same energies, the hydrogen oscillator must be nearly isotropic and have about the same frequency.[3]

Not all hydrogenous substances demonstrate these chemical binding effects to the degree shown above for zirconium hydride and yttrium hydride. Figure 5 shows the relative hydrogen cross section for a powder sample of magnesium hydride. The attenuation of the quartz sample holder has been taken into account. The lighter mass of the magnesium atom compared to zirconium gives rise to a substantial modification of the cross section due to the thermal motion. Figure 2 shows, nevertheless, that this effect would still leave the peaks readily recognizable for magnesium hydride if it has only one system of energy levels as in yttrium hydride and zirconium hydride. Since magnesium hydride has

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[3] Separate measurements to be published elsewhere have shown that the energy level in yttrium hydride is slightly less than in zirconium hydride.

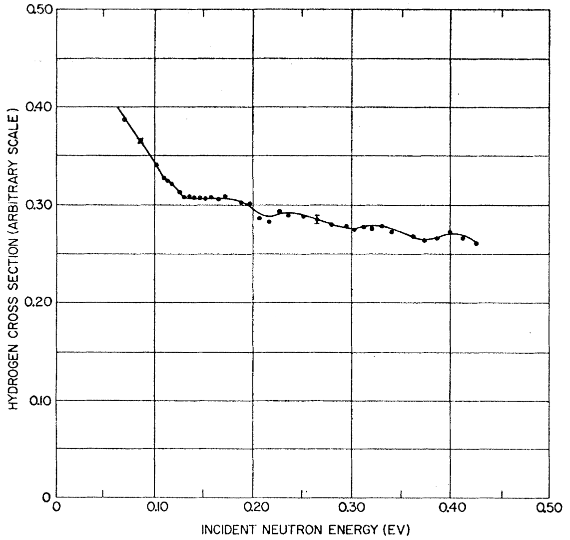

FIG. 5. Total neutron cross section for hydrogen in magnesium hydride (MgH_2), plotted on an arbitrary scale, with errors as indicated.

a layered structure, however, more than one oscillation frequency should be involved, and the resulting curve can crudely be considered as the superposition of at least two curves of the general type shown for zirconium hydride with the minima for each curve occurring at different energies. This effect, coupled with the increased thermal motion due to the lighter mass of the magnesium, would be expected to eliminate the sharp maxima and minima in total cross section observed for isotropic chemical binding. Indeed, although Fig. 5 shows clear evidence of chemical binding effects on the cross section of hydrogen in magnesium hydride, there are no clearly resolved energy levels as found in the other cases. As mentioned above, a measurement was also made on water with the high-resolution spectrometer to see if any structure in the curve not previously reported would be revealed. The results were found to be in good agreement with the smoothly varying cross section already reported in the literature. This would mean that the water molecule has no discrete energy levels in this energy region, in agreement with the results published elsewhere[4] of a detailed study of the chemical binding of hydrogen in water.

The original theory of Fermi as modified to take account of the finite masses of the atoms is nearly adequate to describe the observed variation of the hydrogen cross section with incident neutron energy. There are discrepancies, however, which are probably due to an oversimplification in the model. A more detailed computation taking into account the Debye distribution of the phonons in the crystal would probably give a better description of the cross section in the region where there are now discrepancies. For the hydrides having an isotropic potential well for the hydrogen, this type of measurement can determine the harmonic energy level with an accuracy of about 5% If the potential well is anisotropic (as in the case of magnesium hydride) or involves possibly several types of harmonic motion (as in the case of water), then the resulting curve for the hydrogen cross section is too complicated to give detailed information on the chemical binding of the hydrogen atom.

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[4] A. W. McReynolds *et al.*, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), p. 1540.