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NEUTRON INTERACTIONS IN ZIRCONIUM HYDRIDE

by

W. L. Whittemore

January 6, 1964

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The work reported herein
was supported by the U. S.
Atomic Energy Commission
under Contract AT(04-3)-167
Project Agreement No. 10
General Atomic Project No. 220

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ABSTRACT

Earlier measurements of neutron interactions in zirconium hydride made under this program are reviewed and combined with the latest results obtained with the General Atomic neutron velocity selector. Recent measurements have included (1) the angular dependence of purely elastic scattering from hydrogen atoms in zirconium hydride, (2) the angular dependence of scattering from the first and second excited level, (3) the width of the first and second excited levels, and (4) the temperature and angular dependence of the level width. These results are compared with predictions for a harmonic oscillator. The significant deviations indicate that the potential well for the hydrogen atom differs appreciably from purely harmonic. The temperature and angular dependence of the width of the first excited level clearly indicate that the level width is mainly due to causes other than Doppler broadening.

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I. INTRODUCTION

Zirconium hydride is an example of a neutron moderator with rather unique properties. Instead of a large and overlapping continuum of energy exchange mechanisms, ZrH has unique and well-separated energy levels. Since the hydrogen atom is bound within a tetrahedron of zirconium atoms, in the first approximation the hydrogen is an isotropic, harmonic oscillator. In the present program, the first measurements of neutron scattering in ZrH were made to verify the general features of hydrogen binding. The total scattering cross section and the inelastic scattering of cold neutrons substantiated the concept of a hydrogen atom bound with distinct oscillator levels. Subsequently, other measurements were made in other laboratories to specify, in somewhat greater detail, the exact nature of the binding of hydrogen in ZrH. The most detailed of these were performed by Brockhouse.⁽¹⁾ Although his measurements on the first level substantiated our earlier results obtained from the scattering of cold neutrons, his measurements on the second level disagreed with our earlier published results and indicated a much broader width.

With the advent of our present neutron velocity selector, which provides monoenergetic neutrons with energies up to 0.4 ev, a suitable technique became available for investigating the interaction of incident high energy neutrons on ZrH. Some of the results of this investigation are presented in the following subsections.

II. PREVIOUS EXPERIMENTAL RESULTS ON THIS PROGRAM

2.1 TOTAL SCATTERING CROSS SECTION

Fermi⁽²⁾ was one of the first to point out that the total scattering cross section will reflect specific details of harmonic binding of hydrogen atoms in a crystal. Figure 1 shows a theoretical representation of the notions presented by Fermi. It is seen that for multiples of the harmonic frequency $h\nu$, sharp minima appear in the total cross section. A precision measurement of the total cross section in ZrH was made, using a Brookhaven neutron spectrometer, with results as shown in Fig. 2. An analysis of these experimental data in terms of the theory of Fermi gives a value for $h\nu$ of 0.137 ev for the oscillator level in ZrH. A further examination of Fig. 2 indicates that the theory and experiment deviate in some details around the first and second oscillator levels in ZrH. The deviation is

Fig. 1--Theoretical neutron cross section of bound proton, with frequency ν of isotropic harmonic oscillation.

Fig. 2--The solid curve gives the total neutron cross section of hydrogen in $\text{ZrH}_{1.5}$ obtained from total cross section by subtracting the constant cross section of zirconium. The Fermi theoretical curve corrected for Doppler broadening 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.

greater for the second level, indicating that the isotropic harmonic oscillator model used for the theoretical curve is not completely accurate.

2.2 SCATTERING OF LOW ENERGY NEUTRONS BY ZIRCONIUM HYDRIDE

Previous work⁽³⁾ has shown that the total neutron cross section of hydrogen in ZrH depends significantly on the energy of the incident neutron. For incident neutrons with lower energy ($<h\nu$), one expects that, as the incident energy is decreased, the cross section of hydrogen bound as a harmonic oscillator should, to the first approximation, increase to the fully bound hydrogen cross section of 80.4 barns. The slow increase of the total cross section above the value for a fully bound hydrogen atom is due to inelastic scattering from the acoustical modes and is determined by the frequency distribution in the solid.

To compute the cross section of zirconium hydride, we have used the results of Marshall and Stuart⁽⁴⁾ with appropriate modifications for the case of a bound hydrogen atom and assumed a Debye frequency distribution for the acoustical modes. For this computation, there are three factors to be considered: (1) a Debye-Waller factor due to optical vibrations; (2) inelastic scattering from acoustical modes; and (3) diffraction effects in zirconium. The latter are small and can be approximately corrected for by subtracting the observed elastic cross section for zirconium for neutron energies above the Bragg cutoff. On this basis, we derived the following expression for the total cross section σ :

$$\sigma = \sigma_b f\left(\frac{E}{h\nu}\right) \left[1 + A_1(x, t) \frac{m}{M} + A_2(x, t) \left(\frac{m}{M}\right)^2 + A_3(x, t) \left(\frac{m}{M}\right)^3 + \dots \right] \quad (1)$$

where

σ_b = 81.4 barns/H atom (bound cross section)

$x = (E/\theta)^{\frac{1}{2}}$

$t = T/\theta$

θ = Debye temperature

E = incident neutron energy

T = temperature of scatterer

m = neutron mass (1)

M = Effective mass of hydrogen atom in the lattice. (It is taken as 91 in this calculation, but a mass of 200 is probably better.)

$$f(E/h\nu) = \left(1 - e^{-4E/h\nu} \right) \frac{4E}{h\nu}$$

$h\nu$ = energy of Einstein level. (0.13 ev was used for ZrH in this calculation; however, more recent measurements indicate that 0.140 ev is a better value.)

The parameters A_1 , A_2 , A_3 in this expression have been computed and tabulated by Marshall and Stuart.⁽⁴⁾

During the measurement, the sample was kept at room temperature. The only remaining parameter subject to choice is the Debye temperature to be assumed for the ZrH lattice. The cross sections have been computed for three different assumptions of reasonable Debye temperatures: 197°K (0.0170 ev), 296°K (0.0255 ev), and 370°K (0.0319 ev). A comparison of theoretical and experimental results (see Fig. 3) shows that they are in quite good qualitative and quantitative agreement, in regard both to the shape and to the absolute magnitude of the curve. However, the predicted cross section is found to be relatively insensitive to the choice of Debye temperature for the lattice; consequently, the agreement between experiment and theory may be regarded as real rather than resulting from a fortuitous choice of Debye frequency.

2.3 COLD NEUTRON INVESTIGATION OF INELASTIC SCATTERING IN ZIRCONIUM HYDRIDE

Cold neutrons from a beryllium-filtered reactor source have been used to study the inelastic scattering of neutrons by ZrH. We have published⁽⁵⁾ some experimental results from such a study. Figure 4 shows the effect of temperature on the scattering from hydrogen in ZrH. In general, cold neutron scattering from bound levels as high as those in ZrH is difficult to achieve by the energy gain technique because of the very small population of these higher-lying levels due to the Boltzmann population factor $e^{-h\nu/kT}$. One saving feature for this scattering is that the cross section for energy gain is proportional to $(E/E_0)^{1/2}$ where, for cold neutron experiments, E , the final energy, is of the order of 0.14 ev and E_0 is of the order of 0.003 ev. This feature is insufficient to compensate for the Boltzmann factor, however, and the higher lying levels in ZrH can be observed only if the specimen is heated. Figure 4 shows that for room temperatures, the second level in ZrH does not give any appreciable scattering. Raising the temperature to about 693°K raises the population of both the first and second levels and makes possible the observation of scattering from the second level. By studying the change in population of the first level as a function of temperature, it was possible to evaluate $h\nu$ through the use of the Boltzmann factor $e^{-h\nu/kT}$. This particular measurement gave a value of 0.130 ± 0.005 ev. This value is somewhat lower than the value 0.137 ev obtained from analysis of the total scattering cross section.

Fig. 3--Cross section of H in ZrH at low energy

Fig. 4--Scattering of cold neutrons at $\theta = 90$ deg by $\text{ZrH}_{1.6}$. Note that the width of the first bound level in ZrH increases with temperature and that the second bound level has an intensity which is difficult to measure at room temperature.

The variation of the width of the first and second bound level in ZrH as a function of scattering angle is an indicator of the nature of binding. Earlier in this program, observations were made only at 90 deg; therefore, no firm conclusion could be reached concerning the nature of the binding. However, from the data in Fig. 14, we observe that the width at half maximum of the first harmonic level for ZrH at room temperature is of the order of 0.030 ev. Raising the temperature to about 636°K gives a width for this level of about 0.04 ev. For this elevated temperature, the width of the second harmonic level is about 0.065 ev. From these data, two aspects of the binding in hydrogen can be investigated. The ratio of the widths at half maximum of the second level and the first level is about 1.6. A ratio of $\sqrt{2}$ would be expected if the levels were purely harmonic. Within the accuracy of the experiment, it appeared that the width is somewhat greater for the second level than would be expected for harmonic lattice vibrations.

One can compare the widths of the first level as a function of temperature. The width of the level varies as $(\kappa^2 \bar{T}/M)^{1/2}$ where κ is the magnitude of the momentum transfer, M is the effective mass for the acoustic vibrations of the H atoms (≈ 200), and $(3/2)\bar{T}$ is the average kinetic energy of the hydrogen atoms in a Debye crystal. $\bar{T}$ is given by the expression:

$$\bar{T} = \frac{1}{2} \int \omega \rho(\omega) \coth\left(\frac{\omega}{2T}\right) d\omega, \quad (2)$$

where T is the temperature in ev units. Making a high temperature expansion where $\omega/2T$ is small, one finds

$$\bar{T} = T + \frac{\theta^2}{20T}, \quad (3)$$

where θ is the Debye temperature in the same units as T and is about 300°K. From the above, the ratio of the level widths at 300°K and 636°K is predicted to be 1.43. Experimentally, the ratio of the two widths at half maximum is of the order of 1.33, which is in reasonably good agreement with the ratio predicted for pure Doppler broadening.

Considering the observed width of ≈ 0.030 ev at room temperature, we can compute the expected width of the optical band due to the finite mass of the zirconium atoms. Since the relative inherent width $\Delta\omega/\omega$, given by $(1 + \frac{m}{M})^2 - 1$, is of the order of 0.005 or smaller, depending on whether M is taken as 91 or larger, the inherent line width is of the order of $0.005 \times 0.140 \approx 0.0007$ ev. This is far smaller than the 0.030 ev measured above and indicates that the width is not due to the finite mass of zirconium atoms.

III. INELASTIC SCATTERING FROM ZIRCONIUM HYDRIDE USING ENERGY-LOSS TECHNIQUES

From Section 2.3, it is clear that the full range of studies of neutrons interacting with ZrH can be carried out only when high energy incident neutrons are available, because of the effect of the Boltzmann population factor in populating mainly the lowest lying level at room temperature. The present neutron velocity selector provides monoenergetic neutrons with energies up to 0.4 ev. These monoenergetic beams have now been used to study additional characteristics of the binding of hydrogen in ZrH. Since the harmonic frequency in ZrH is of the order of 0.14 ev, these neutrons can be used to study directly the characteristics of the second bound level in ZrH using energy-loss techniques. It will be remembered that the differential inelastic scattering cross section for neutron gain experiments has a factor of $(E/E_0)^{\frac{1}{2}}$ which represents an advantage for energy gain scattering and some disadvantage for energy loss scattering. However, the Boltzmann population factor worked severely against neutron-energy-gain scattering but is not important for energy loss experiments.

3.1 SCATTERING PATTERNS FROM ZIRCONIUM HYDRIDE - EXPERIMENTAL RESULTS

Scattering patterns have been obtained for a variety of incident neutron energies and scattering angles by utilizing the neutron velocity selector to provide monoenergetic incident neutrons. Some of the experimental conditions were set up to optimize conditions for examining characteristics of the first or second harmonic level in ZrH. Other scattering patterns were selected to provide general information for the derivation of the generalized Scattering Law representation of neutron interactions in ZrH.

The selection of optimum conditions for studying the first or second scattering level in ZrH requires attention to multiple scattering effects. From the information shown in Fig. 2, one sees that for neutron energies greater than about 0.09 ev, the cross section of hydrogen bound in ZrH is roughly constant in the range of 20-25 barn. For neutron energies below 0.09 ev, the cross section rises rapidly to much higher values. This fact alone creates a considerable problem in the selection of sample thickness. If an experiment is designed to minimize multiple scattering, this consideration is particularly important. For instance, if incident neutron energies of the order of 0.240 ev are selected and allowed to scatter on a relatively thin ($T = 0.9$) sample of ZrH, the neutron which has excited the first harmonic level will emerge from the scattering sample with an energy centered around 0.10 ev. The cross section for such a neutron upon leaving the scattering sample is no larger than for the incident neutrons. On the other hand, if an incident energy of about 0.20 ev is

selected for the incident monoenergetic beam and the same inelastic neutron interaction is studied, the outgoing neutron will have an energy centered around 0.06 ev. The multiple scattering effects for such an outgoing neutron are considerably larger than in the former case. If an even smaller incident energy is selected, a situation rapidly develops in which the outgoing neutron has practically no chance of emerging from the ZrH sample without undergoing further scattering. In this regard, we believe that the scattering samples used by Brockhouse⁽¹⁾ were much too thick, since incident energies of the order of 0.15 ev were used with a sample having a transmission of 0.7. The outgoing neutron energy was of the order of 0.003 ev and therefore underwent numerous scattering before emerging from the sample. For the second bound level in ZrH, incident neutron energies of the order of 0.38 ev are selected. The outgoing neutron in these cases also has an energy of the order of 0.10 ev and thereby suffers the same small multiple scattering as before. Figure 5 shows typical time-of-flight results of the neutron scattering by the first bound level in ZrH. Observations were made at 30 deg and 90 deg in a ZrH specimen having a chemical formula $\text{ZrH}_{0.5}$. The first measurements in this series were made with a specimen having a thickness of 0.080 in. This gave a transmission of 0.9 for high energy neutrons where the cross section is of the order of 25 barns. All later measurements in the present series were made with finely powdered $\text{ZrH}_{2.0}$. The thickness of the specimen was held quite closely to 0.020 in., also giving a transmission of the order of 0.9 for high energy neutrons. The observed width of the harmonic level appears to be exactly the same for the hydrogen in these two different concentrations. The incident energy in this particular case is 0.239 ev. In Fig. 6, one finds similar scattering patterns for $\text{ZrH}_{2.0}$ where incident neutron energies of the order of 0.39 ev were selected. Here one sees clear evidence for excitation of the first and second harmonic levels. These data in both Figs. 5 and 6 are particularly useful for studying several aspects of the binding of hydrogen in ZrH. In Fig. 7, we have shown, for comparison purposes, the scattering patterns observed at three scattering angles for a monoenergetic incident neutron beam having selected energies too low to excite the first hydrogen oscillator level. The difference in the scattering pattern for the lower incident energy, which is readily apparent, is directly related to the ability of ZrH to act as an exceedingly fine neutron moderator for incident neutron energies above the energy of the first harmonic level.

3.2 SOME CONSIDERATIONS OF THE NATURE OF HYDROGEN BINDING IN ZIRCONIUM HYDRIDE

Because hydrogen is bound in a nearly equilateral tetrahedron of Zr atoms and because the total cross section has been shown to reveal some of the characteristics of harmonic oscillator binding, it seems reasonable to compute some of the theoretical predictions of the expected behavior of

Fig. 5--Time-of-flight scattering data for $\text{ZrH}_{0.5}$ with and without background for 300°K at $\theta = 30$ and 90° . $E_0 = 0.237 \pm 0.006 \text{ eV}$. No noticeable difference in the width of the scattered distribution is observed for $\text{ZrH}_{2.0}$.

Fig. 6--Time-of-flight scattering data for $\text{ZrH}_{2.0}$ for 300°K at $\theta = 30, 60$, and 90 deg.
 $E_0 = 0.386 \text{ eV}$. The data are shown with background subtracted.

Fig. 7--Time-of-flight scattering data for $\text{ZrH}_{2.0}$ with background deducted for 300°K at $\theta = 30^\circ, 60^\circ$, and 90° , respectively. Incident neutron energy ($E_0 = 0.10 \text{ eV}$) was selected to be too low to excite the first vibrational level of hydrogen. Note that the wings of the quasi-elastic scattering are considerably broader than for the data taken with higher E_0 as in Figs. 4 and 5.

a purely harmonic oscillator. The results of these predictions will then be compared directly with experiments, in an effort to ascertain the degree to which ZrH satisfies the assumptions of an isotropic, harmonic oscillator. From a multiphonon expansion, one can write the differential energy-loss cross sections for the first harmonic and second harmonic levels as follows:

$$\frac{d^2\sigma_1}{dE d\Omega} = \frac{\sigma_b}{4\pi} \left(\frac{E}{E_0} \right)^{\frac{1}{2}} \exp\left(-\frac{\kappa^2 \alpha}{2}\right) \exp\left(\frac{\omega_0}{2T}\right) I_1 \left[\frac{\kappa^2}{2\omega_0 \sinh(\omega_0/2T)} \right] \delta[(E_0 - E) - \omega_0] \quad (4)$$

$$\frac{d^2\sigma_2}{dE d\Omega} = \frac{\sigma_b}{4\pi} \left(\frac{E}{E_0} \right)^{\frac{1}{2}} \exp\left(-\frac{\kappa^2 \alpha}{2}\right) \exp\left(\frac{\omega_0}{T}\right) I_2 \left[\frac{\kappa^2}{2\omega_0 \sinh(\omega_0/2T)} \right] \delta[(E_0 - E) - 2\omega_0] \quad (5)$$

where

σ_b = the bound cross section = 80 barns

E, E_0 = the final and incident energy, respectively

$\frac{\kappa^2}{2} = E_0 + E - 2(E_0 E)^{\frac{1}{2}} \cos \theta$, where θ is the scattering angle

$$\alpha = \frac{2\bar{n}+1}{\omega_0} ; \quad \bar{n} = \left[\exp(\omega_0/T) - 1 \right]^{-1}$$

ω_0 = oscillator level in ev

T = sample temperature in ev

$$I_n(x)_{x \rightarrow 0} = \frac{1}{n!} x^n$$

In Figs. 8 and 9, we have shown, as a function of angle between the limits of 30 deg and 120 deg, the theoretical values of the cross section integrated over the energy range for the first and second harmonic levels. Figure 10 shows the theoretical angular dependence of the first harmonic level when the incident neutron energy is 0.239 ev, for which energy it is not possible to excite the second harmonic level.

From such experimental data as are contained in Figs. 15 and 16, it is possible to make the intercomparison of the first and second harmonic level in a number of ways. One can confine attention to a single level (e.g., the first or second excited level) and study its variation as a

Fig. 8--Theoretical plot of the angular distribution for the partial cross sections for scattering from the first and second bound hydrogen levels in ZrH. $E_0 = 0.380$ eV. A multi-phonon scattering treatment was used.

Fig. 9--Theoretical angular distribution for scattering from the first and second level in ZrH. $E_0 = 0.335$ ev. A multi-phonon scattering treatment was used.

Fig. 10--Theoretical angular distribution for scattering from the first excited level in ZrH. $E_0 = 0.239 \text{ eV}$. The second level cannot be excited with this incident energy. A multi-phonon scattering treatment was used.

function of angle or one can fix the observation angle and study the intensity ratio of the first to the second level. Both of these approaches have been made for the existing data. Table 1 presents this information as a function of scattering angle for $E_0 = 0.335$ ev and 0.386 ev. Table 2 presents the information obtained by comparing the intensity ratio of the first and second levels for fixed observation angles. The information in Tables 1 and 2 indicates that the binding of hydrogen in $ZrH_{2.0}$ differs from that expected for a purely harmonic crystal. Discrepancies involving factors of three or four are observed in Table 2 and are considered significant. Specifically, the angular dependence of the scattering from the first level is about as expected for a harmonic potential. However, the deviations for the second level are very large.

Table 1

ANGULAR VARIATION OF PARTIAL CROSS SECTION
FOR FIRST AND SECOND LEVEL, $d\sigma_1/d\Omega$ AND $d\sigma_2/d\Omega$
RESPECTIVELY: THE DATA ARE NORMALIZED
TO UNITY AT 30 DEG.

E_0 (ev)	θ (deg)	$d\sigma_1/d\Omega$		$d\sigma_2/d\Omega$	
		Theoretical	Exp.	Theoretical	Exp.
0.335	30	1.00	1.00 ± 0.045	1.00	1.00 ± 0.045
	60	0.78	- - -	2.354	- - -
	90	0.25	0.34 ± 0.02	1.53	1.29 ± 0.05
0.386	30	1.00	1.00 ± 0.04	1.00	1.00 ± 0.04
	60	0.67	0.66 ± 0.03	2.20	1.34 ± 0.06
	90	0.15	0.169 ± 0.007	0.95	0.79 ± 0.03

Table 2

RATIO OF THE PARTIAL CROSS SECTION FOR THE
FIRST $\sigma(\omega_0)$ AND SECOND $\sigma(2\omega_0)$ EXCITATION
LEVEL IN ZIRCONIUM HYDRIDE: THE THEORETICAL
VALUES ARE EVALUATED FOR A HARMONIC OSCILLATOR

E_0 (ev)	θ (deg)	$\sigma(\omega_0)/\sigma(2\omega_0)$ Theoretical	$\sigma(\omega_0)/\sigma(2\omega_0)$ Experimental
.335	30	1.60	2.90 ± 0.08
	60	0.53	- - - -
	90	0.26	0.77 ± 0.02
.386	30	1.44	2.63 ± 0.06
	60	0.44	1.31 ± 0.03
	90	0.23	0.56 ± 0.02

The purely elastic scattering (where the incident and outgoing energies are exactly the same) also provides a test for variations from pure harmonic binding. If the vibrations are strictly harmonic, even through not isotropic, the differential angular scattering of this purely elastic scattering $\frac{d^2\sigma}{dE d\Omega} \big|_{el}$ is given as follows:

$$\frac{d^2\sigma}{dE d\Omega} \big|_{el} = \frac{\sigma_b}{4\pi} \exp(-\kappa^2 \alpha / 2) \delta(E_0 - E) , \quad (6)$$

where σ_b , κ^2 , α , E_0 , and E have been previously defined. Using the fact that $E_0 = E$ and integrating over all values of E , we find that Eq. (6) becomes

$$\frac{d\sigma}{d\Omega} \big|_{el} = \frac{\sigma_b}{4\pi} \exp(-4E_0 \alpha \sin^2 \frac{\theta}{2}) \quad (7)$$

From Eq. (7), one sees that the integrated intensity of pure elastic scattering should have a simple angular dependence given essentially as

$$\ln \frac{d\sigma}{d\Omega} \big|_{el} = K - 4E_0 \alpha \sin^2 \frac{\theta}{2} , \quad (8)$$

where K is a constant. Thus, a plot on semilog paper of the scattered

elastic intensity should give a linear dependence on $4E_0 \sin^2(\theta/2)$ if the potential well is harmonic.

Figures 11a and 11b show the experimental scattering data for the elastic peaks for samples of $ZrH_{0.5}$ and $ZrH_{2.0}$. The data plotted in Figure 11a for $ZrH_{2.0}$ are from the same set as that which was used to investigate the inelastic scattering above. The scattering contribution due to the zirconium atoms and the aluminum holder have been deducted by using as a standard the scattering produced by a specimen of vanadium. These data show a significant deviation from linearity for large momentum transfer. Unfortunately, the correction due to the scattering from the aluminum holder is large compared with that from the hydrogen atoms since a thick holder was necessary to maintain a uniform thin thickness of the powdered $ZrH_{2.0}$. A small error in this larger contribution may have caused the resulting scattering from the hydrogen atoms to deviate greatly from an otherwise linear dependence on $4E_0 \sin^2(\theta/2)$. In order to check more closely on this linearity, a separate experiment was performed with a metallic specimen of $ZrH_{0.5}$ held in an aluminum holder reduced in total thickness from about 0.25 in. to about 0.050 in. The results are shown in Fig. 11b. The scatter of the data points is due to the poorer statistics caused, in part, by the lowered concentration of hydrogen. However, the new data exhibit an essentially linear dependence on $4E_0 \sin^2(\theta/2)$ within statistics.

At the present time, one must conclude that within the accuracy of the experiment the elastic scattering from $ZrH_{0.5}$ gives no evidence for anharmonic motion of the hydrogen atoms. For a specimen of $ZrH_{2.0}$, the conclusion is not certain since the deviation from a linear dependence on $4E_0 \sin^2(\theta/2)$ may be due either to anharmonic motion of the hydrogen or to an improper background determination. From the theoretical viewpoint, one might reasonably expect the purely elastic scattering to vary in the manner prescribed for a harmonic oscillator in the ground state and to exhibit deviations from harmonic behavior for the higher bound levels where the potential well may begin to deviate from the harmonic shape.

The width of the energy distribution of scattered neutron is of interest in the determination of the nature of hydrogen binding in ZrH . From Section 2.3, we have already shown that cold-neutron energy-gain scattering has given a scattering width at half maximum of the order of 0.030 eV for the first level and is far larger than the inherent line width due to the finite mass of the zirconium atoms. The widths of the excitation levels are evaluated for the experimental data of Figs. 5 and 6 and are given in Fig. 12. From Fig. 12, one obtains an observed line width of 0.033 eV and 0.055 eV respectively. At present, an extensive investigation of the instrumental resolution for $ZrH_{0.5}$ is under way.

Fig. 11a--The dependence of elastic scattering on $4E_0 \sin^2(\theta/2)$, the momentum transfer for $\text{ZrH}_{2.0}$. The deviation from the straight line for $4E_0 \sin^2(\theta/2) > 0.7$ indicates anharmonic motion of hydrogen atoms.

Fig. 11b--The dependence of elastic scattering on $4E_0 \sin^2(\theta/2)$, the momentum transfer for $\text{ZrH}_{0.5}$. The data are consistent with a straight line and hence indicate harmonic motion of the hydrogen atoms.

Fig. 12--Distribution in final energy for scattering from the first and second bound levels of H in ZrH . Two values of E_0 were chosen; i.e., 0.239 eV and 0.386 eV. The indicated width at half maximum includes instrumental resolution as well as the inherent line width of the first and second bound levels.

In the future, a better evaluation of the resolution effects on the data will be possible. At this writing, the energy resolution for the first level is known to be quite close to 0.020 ev, whereas that for the second-level measurements is believed to be worse, ranging between the limit 0.025 ev and 0.040 ev. Thus, the true width of the distribution of scattered neutrons due to the line width is close to 0.025 ev for the first level, but for the second level we can only specify that the level width lies in the range 0.30 ev to 0.045 ev. If the potential well is harmonic, the second level width should be $\sqrt{2}$ times the first level width and thus would be about 0.035 ev. Anharmonic binding effects would broaden the second level. The uncertainty in the width of the second level makes any firm conclusion on the shape of the potential well unlikely from the level width data.

For a one-phonon theoretical treatment of the scattering due to the first bound level, one finds that the frequency distribution $\rho(E_0 - E)$ is given as:

$$\begin{aligned} \frac{d^2\sigma}{dE d\Omega} &= A \left(\frac{E}{E_0} \right)^{\frac{1}{2}} \exp(-\kappa^2 \alpha / 2) \frac{\kappa^2 \alpha}{2} \frac{\rho(E_0 - E) \exp[-(E_0 - E)/2T]}{2(E_0 - E) \sinh[(E_0 - E)/2T]} \\ &= A B(E_0, E, \theta) \rho(E_0 - E) \end{aligned} \quad (9)$$

where

A = a proportionality constant

$\hbar = k = 1$

$E_0 > E$, and

κ^2 , E_0 , E , α , and T are as previously defined.

One can evaluate the frequency distribution for zirconium hydride from experimental distributions, such as are given in Fig. 12, using the kinematic corrections contained in $B(E_0, E, \theta)$. Figure 13 shows the magnitude of this correction. The resulting frequency distribution for the first excited level in zirconium hydride is given in Fig. 14. Since the instrumental resolution is independently evaluated to be 0.023 ev, the observed width for $\rho(\omega)$ at half maximum of 0.036 ev gives a true width in the frequency distribution of 0.028 ev.

One parameter of considerable interest in reactor calculations is the quantum $\hbar\nu$ for the hydrogen oscillator. As noted in Section II, our previous values have ranged between 0.130 ev $\pm$ 0.004 ev and 0.137 ev. Brockhouse⁽¹⁾ has reported a value of 0.140 ev for his energy-loss scattering measurements. The present series of experiments give values of $\hbar\nu(1)$ in the range 0.140 ev to 0.145 ev, depending on whether the peak in frequency distribution (0.145 ev) or in the energy distribution (0.140 ev)

Fig. 13--Typical set of correction terms $B(E_o, E, \theta, \omega_o)$ to be used to derive a frequency distribution $\rho(\omega)$ for ZrH. E_o was taken as 0.239 ev and ω_o was taken as 0.140 ev.

Fig. 14--Experimentally determined frequency distribution $\rho(\omega)$ for ZrH. $E_0 = 0.239 \text{ ev}$. This curve includes a contribution due to instrumental resolution; note, however, that the width at half maximum is broader than the distribution in final energy shown in Fig. 12.

of scattered neutrons is used. In considering the above data, we conclude that the value of 0.130 ev is somewhat too low and that a value of about 0.140 ev is more nearly correct. The value of $h\nu(2)$, determined from the peak of the distribution of neutrons scattered by the second bound level of hydrogen, appears to be 0.285 ev for $E_0 = 0.335$ ev, and 0.291 ev for $E_0 = 0.386$ ev. Since the distributions are quite broad, it is difficult to pick a unique, meaningful value. Consequently, we cannot determine whether the second level $h\nu(2)$ is a simple integral multiple of the first level $h\nu(1)$, although there is some slight evidence that the ratio is slightly larger than 2.0.

A measurement was made of the scattering from the first bound level of ZrH as a function of the temperature of the specimen. The incident neutron energy was 0.239 ev. The two temperatures of the sample were 300°K and 90°K. Figure 15 shows that, in the time-of-flight distributions, a real decrease in line width occurs for this change in ambient temperature. Similar data exist at other scattering angles. It should be noted that Brockhouse⁽¹⁾ has published results which indicate no change in line width, within his accuracy, for a similar change in temperature. The reasonable conjecture is that Brockhouse's sample thickness was great enough to give much multiple scattering and thus masked the relatively small change in width caused by a change in temperature. In this same connection, it may be noted that the line width reported by Brockhouse for the second bound level is more than a factor of two wider than found in the present experiments, probably for the same reason.

For the present measurements, we have calculated that the corrected line width of the first scattered level is 0.025 ev for a temperature of 300°K. The data in Fig. 15 give a line width of 0.020 ev after resolution corrections for $T = 90^\circ\text{K}$. The ratio of these line widths is about 1.25. The predicted decrease in width due to Doppler broadening effects is obtained from $(\kappa^2 \bar{T}/M)^{1/2}$, where $\bar{T}$ is defined by Eqs. (2) and (3). Equation (3) is valid within 5 percent for all $\bar{T}$ greater than $\theta/4$. From Eq. (3), we note that the ratio of $\bar{T}$ for these two temperatures gives a value of 2.25. Thus the ratio of the resulting level widths at 300°K and 90°K is 1.50, much larger than the observed experimental value of 1.23. However, the magnitude of M above is believed to be so large as to make the magnitude of the predicted level width much too small due to these Doppler effects. Furthermore, it should be noted that a Doppler broadening of the level will also introduce an angular dependence for the line width because of the momentum transfer. For the present case, with $E_0 = 0.239$ ev, $E = 0.10$ ev, and $\theta = 30$ deg and 90 deg, one predicts that the line width should increase by a factor of 2.2 for scattering at 90 deg compared to 30 deg. Experimentally, one observes that the line width is practically independent of the scattering angle (from 30 deg to 90 deg) for $T = 90^\circ\text{K}$ and increases only slightly (≈ 10 percent) with angle for $T = 300^\circ\text{K}$. All of these observations are at variance with

Fig. 15--Experimental distribution in time-of-flight for scattering from the first bound level in ZrH shown as a function of sample temperature. Note that the width at half maximum is noticeably narrower for the lower temperature of 90°K.

those predicted on the basis of simple Doppler broadening of the optical band. A more detailed consideration of the exact phonon distribution is needed for comparison with the present experimental data.

3.3 A SUMMARY OF EXPERIMENTAL RESULTS AND CONCLUSIONS ON SCATTERING OF NEUTRONS BY ZIRCONIUM HYDRIDE

1. $h\nu(1) = 0.140$ to 0.145 ev.
 $h\nu(2) = 0.285$ to 0.291 ev.
2. In the temperature range from 300°K to 690°K , the width of the first bound level is observed to change with temperature nearly as predicted by $(\kappa^2 \bar{T}/M)^{\frac{1}{2}}$, where κ is the magnitude of the momentum transfer and $\bar{T}$ is related to the average energy of an H atom in the Debye crystal. In the temperature range between 90° and 300°K , the width of the first bound level is observed to change by a factor of 1.25 whereas a Doppler-broadened level would be expected to change its width by a factor of 1.5.
3. A true Doppler broadening of the level also requires an angular variation of the level width. This is nearly absent at 90°K in the present sample for angles in the range of 30 deg to 90 deg and much smaller at $T = 300^\circ\text{K}$ than the predicted factor of 2.2.
4. The natural line width due to the finite mass of the zirconium is 0.0007 ev or less, whereas the observed width at 300°K at half maximum of the distribution of scattered neutrons is 0.025 ev. The observed width for the second level is uncertain within the limits of 0.030 and 0.050 ev. It is thus not certain whether the ratio of the level widths is $\sqrt{2}$ as is expected for harmonic lattice vibrations.
5. The calculated experimental half width of the frequency distribution $\rho(\omega)$ for the first level is 0.028 ev for $T = 300^\circ\text{K}$.
6. The cross sections for scattering from the first level vary with scattering angle as predicted for a harmonic oscillator. The angular variation in scattering from the second excited level is quite different from that predicted for a harmonic oscillator. Differences of a factor of three are observed.
7. The cross section for purely elastic scattering by hydrogen atoms bound in $\text{ZrH}_{0.5}$ varies linearly with momentum transfer (up to the maximum investigated) as required for harmonic motion. For $\text{ZrH}_{2.0}$, the variation is linear for the smaller momentum transfers but appears to deviate from linearity for larger momentum transfers, indicating the possibility of a difference in the binding of hydrogen atoms in $\text{ZrH}_{0.5}$ and $\text{ZrH}_{2.0}$.

8. Items (1), (4), and (5) are consistent with pure harmonic motion of the hydrogen but do not necessarily rule out anharmonic and/or anisotropic motion. On the other hand, item (6) is inconsistent with harmonic motion and indicates anharmonic binding for the second excited level of the hydrogen in ZrH. Item (7) is ambiguous as regards the harmonic nature of the hydrogen motion inasmuch as the results for $\text{ZrH}_{2.0}$ indicate anharmonic motion, whereas measurements on $\text{ZrH}_{0.5}$ with more attention to background corrections indicate harmonic motion of the hydrogen.

IV. ACKNOWLEDGEMENTS

The writer wishes to acknowledge profitable discussions with D. E. Parks and M. S. Nelkin. Appreciation is expressed for the assistance of the operating staff of the General Atomic electron linear accelerator. Special acknowledgement is hereby expressed for the assistance given by Arthur Hom in operating the apparatus and reducing the data.

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