Neutron and X-ray School 2016
Oak Ridge/ Argonne

ARCS Experiment Procedure
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Dynamics of metal hydride systems: Harmonic oscillators and beyond
or
Is ZrH_2 a collection of quantum harmonic oscillators?

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Experiment Summary

The hydrogen in zirconium hydride (ZrH_2) sits at the interstitial positions between the zirconium. At the simplest description, the energy levels can be considered to be the same as a particle in a potential well. The aim of this experiment is to measure the vibrational spectrum of ZrH_2 as a function of energy and wavevector transfer, and determine how well it conforms to the predictions of the scattering from quantum harmonic oscillators.

The experiment will consist of mounting the ZrH_2 sample in the instrument and starting the data acquisition system to collect the high energy excitation spectrum. An initial analysis of the time-of-flight data will be done to get an overview of the technique. More detailed data reduction will follow using reduced data, leading to more quantitative analysis of the momentum transfer dependence for different energy cuts of the scattering function $S(Q,\omega)$.

If time permits, the group may continue measurements with other instrument settings, or try a different system that displays the quantum harmonic oscillator behavior.

Acknowledgement This experiment follows the tradition of previous neutron and x-ray schools at Argonne National Laboratory, where ZrH_2 was studies on LRMECS at the Intense Pulsed Neutron Source. Ray Osborn and colleagues are thanked for passing on this interesting and illustrative experiment. Thanks to Adam Aczel and collaborators for providing the opportunity to measure UN.

Detailed Procedure with questions and comments

Note: Data file numbers and some parameters to functions will need to be changed to perform the analysis on the data taken by each group. The examples use previously recorded data in run ARCS_51909 saved in experiment /SNS/ARCS/IPTS-9817/ using a working directory /SNS/ARCS/IPTS-2014/shared/ZrH2_testing/.
Students will work in the 2016 NXS proposal directory /SNS/ARCS/IPTS-2014/ with a working directory in /SNS/ARCS/IPTS-2014/shared/.

1. Instrument orientation and data collection

A tour of the instrument will be given by instrument staff. Students work with the staff to mount the ZrH_2 in a sealed aluminum can on the spectrometer and perform the data acquisition. The first measurement will be at 700meV incident energy.

2. Initial setup and analysis of time-of-flight data

Students can split into two working groups using the two ARCS analysis clusters. Subdirectories within the neutron school experimental proposal data storage areas should be made to allow parallel analysis to go on.

Make a directory to keep your work, tagged with your name or another unique identifier. Open a terminal (right click on desktop then "Open in Terminal" or menu item Applications-System-Terminal) and create the directory:

```
$ cd /SNS/ARCS/IPTS-2014/shared/
$ mkdir ZrH2_GroupName
$ cd ZrH2_GroupName
```

Initial viewing of the data will be done with Mantid software. Start a session:

```
$ MantidPlot &
```

Load the datafile you took of the ZrH2 scattering into MantidPlot using the Load-File pull-down located typically in the upper right part of the window labeled Workspaces.
Enter ARCSnnnnn (where nnnnn is your run number) as the file name and wait for the system to find it or Browse to /SNS/ARCS/IPTS-2014/data/ and find the ARCS_nnnnn_event.nxs file. Enter a name to save the data in a workspace, or let the system put in a name like ARCS_nnnnn_event. Check that the box for "LoadMonitors" is set and hit Run.

Now plot the beam monitor data by right clicking the workspace labeled with "monitors" and selecting "Plot Spectrum with Errors…". You want both spectrum 1 (first beam monitor just after the Fermi chopper) and spectrum 2 (second beam monitor in the beamstop). Use the plot controls (typically shown are pan, zoom in, zoom out, rescale all, cursor label, fit) to explore this time-of-flight data to answer the questions below.

<u>Question</u>
What are reasonable times-of-flight corresponding to the 700 meV neutrons at each monitor? Note that you can use the Python window available in MantidPlot for calculations, typically located at the bottom of the window.
FACTS:
~700 meV neutrons
distance from source to mon1 = 11.835 m
distance from source to mon2 = 18.5 m
conversion: E (meV) = 5.23e-6 v(m/s)^2 (in Python exponentiation is **)

Find and note the peak times for each monitor, either by eye or using built-in fitting functions.

(Typically peaktime1 ~ 1020 microseconds and peaktime2 ~ 1600 microseconds.)

Calculate the true incident energy in meV based on the monitor data. Using the Python window:

```
In []: dmon1 = 11.835
In []: dmon2 = 18.5
In []: peaktime1 = 1020
In []: peaktime2 = 1600
In []: vel = (dmon2-dmon1)/(peaktime2 - peaktime1)*1e6
In []: print vel
11491.3793103
In []: E = 5.23e-6*vel**2
In []: print E
690.630905916
```

If you prefer to use a different plotting and analysis package, you can save ASCII data for the monitors using a function in MantidPlot. Highlight the monitors workspace and type SaveAscii in the Algorithms part of MantidPlot. Hit Execute and supply a filename in your working directory to save it.

<u>Question</u>
How does your value compare to the one the Mantid software calculates?
Use the algorithm GetEi with the monitor workspace selected. Use defaults for parameters and supply your own energy estimate to allow the algorithm to find the monitor peaks. Results are displayed in the MantidPlot Results Log.

We can also explore the detector data by plotting it as an image based on the detector array geometry that is stored with the data. Before we start, since the SNS saves event data (each neutron identified by its detector ID and time-of-flight value), we have to tell MantidPlot the range and resolution of data you want to look at.

Select the event data file (ARCS_nnnnn_event) and execute the algorithm Rebin. You can use the same name for the output as the event file (it is really just setting a parameter the program uses), and check the box to PreserveEvents. The parameter can be the bin size you want to keep for viewing the data, so enter 10 in Params and run the rebin algorithm.

Now you can right click on the rebinned event data file and select Show Instrument. This will bring up a view with the data displayed. On the Render tab it is usually most useful to view ARCS data as Full 3D or Cylindrical Y. You can change the data you see with the Time-of-flight bar at the bottom, for example by narrowing the range and scrolling to different times. On the Pick tab you can also see the time-of-flight represented for each pixel (usually very low counts) or by tube (probably more reasonable amount of counts). You can narrow the range plotted by typing values into the boxes to the left and right of the Time-of-flight bar.

<u>Questions</u>
What are the rings that you see in the data summed over all time-of-flight?
What are the different gaps in the array and how might they affect a measurement?
When do the elastically scattered neutrons arrive in time-of-flight, typically the most intense peak in an inelastic time-of-flight spectrum? Is it the same time for all detectors? Why or why not?

What are the neutrons that arrive after the elastic ones?
FACTS:
The sample is 13.6m from the source, and the sample-detector distances vary from 3.0m to about 3.4m.

3. Examining S(Q,E) for the scattering from ZrH_2

In order to analyze the scattering function S(Q,E) that you have measured, we will take advantage of an automatic data reduction process as well as use a different software package. Minimize the MantidPlot window or move to a different screen available for your desktop on the ARCS cluster.

The autoreduction process converts the event-based time-of-flight data to histograms of intensity versus energy transfer for each pixel. The process corrects for detector sensitivity and the solid angle of each pixel using a standard reference run from vanadium (an isotropic incoherent scatter with a well-known cross-section). Data are normalized by the total proton charge sent to the target during the measurement, and scaled by k_i/k_f so that the results are proportional to S(Q,E). The data is saved in a binary format called NXSPE which includes the pixel histograms along with other information needed like the incident neutron energy and the detector geometry.

The program Mslice available in the DAVE software package from NIST can use the NXSPE file to plot your S(Q,E) data. Open DAVE by using the menu Applications-Analysis-DAVE or typing in a terminal:

```
$ dave &
```

Start up Mslice from the DAVE menu Analysis-Mslice TOF/TAS Analysis. If asked select NXSPE as your file type or check it under the menu File-Filetype. Make sure that Mslice is in Powder mode (upper right part of window) and that you are using |Q| and Energy as Viewing Axes. In the data file window (upper left) find your automatically generated reduced data file for the run number of your data, e.g. /SNS/ARCS/IPTS-2014/shared/autoreduce/ARCS_nnnnn_autoreduced.nxspe. Select the file and hit Load Data, accepting defaults of any pop-up windows.

After loading, hit Calculate Projections. This tells the program to use the instrument geometry it just read to precalculate information it needs to display your data in Q and E space.

The first thing to plot will be a Slice, or 2D image view of the intensity. Leaving most fields that control the plot blank will make them defaults to reasonable values. It is recommended to set the step sizes to 0.2 for Q and 5 for E to start. Hit Plot Slice to generate the image of your data. You can change the image by right clicking the color bar to set different intensity ranges or make it logarithmic, using the mouse to zoom in (right clicking in window outside of the image will zoom out), adding text with the Edit menu, etc.

<u>Question</u>
What determines the range in Q that is available? How about energy?
FACTS:
Max angle ~135 degrees

Q (elastic) = 2 k sin(theta)
k = 2 pi/lambda = sqrt(E/2.072) [E in meV and k in inverse Angstroms]

The overall shape of the domain in (Q,E) space in which the instrument has collected data reflects the kinematic constraints from the energy and momentum conservation laws.

Question
Can you identify the features we expect for the quantum harmonic oscillator? Where is the elastic scattering and what are the different energy levels?

The strongest scattering intensity is for the elastic peak (zero energy transfer). Make sure you identify this portion of the data. Stripes at higher energy transfers correspond to excitation of energy levels in the sample by the incident neutrons. The stripes are at constant energies (independent of Q), and explain why there were sharp peaks in I(E) and I(t-o-f) plots. These excitations are called 'dispersionless', as the energy of the excitation does not depend on the momentum. Note how the different excitation energies are multiples of the fundamental.

The image may be saved on the screen for reference by clicking the Keep menu item at top, and stored for later use by the File-Save Plot As- menu item.

It is useful to look at one-dimensional data sets for more details and for future quantitative analysis. Use the Cut tab of the Mslice window to make a 1D plot versus energy by integrating over all of the Q space. You want to Cut along E with an appropriate range and step size while setting the thickness range of Q. Plot Cut will produce the plot, which you may again customize with labels, rescaling, etc.

Once you are satisfied with the data plot, save the data values themselves for future quantitative analysis by using the File-Save Plot As-Save ASCII option. Put the data in your working directory, e.g. /SNS/ARCS/IPTS-2014/shared/ZrH2_GroupName/cutE.txt.

Questions
At this point, without further analysis of the data, what things support the claim that these excitations are due to quantum harmonic oscillators?
Are there aspects that don’t fit that claim?
Hint: What about the resolution variation versus energy transfer for a direct geometry spectrometer.

We will also want to examine in detail the Q dependence of the scattering, so use the Cut tab to make one-dimensional plots along Q by summing over appropriate values of the energy transfer. You want to make versions for the elastic line, say for energies -25 < E < 25meV, with a Q step size of about 0.2 and save it as an ascii file, e.g. cut0.txt. Also save data for different orders of excitations, e.g. cut1.txt, cut2.txt, cut3.txt.

Advanced user note: This version of Mslice has a neat feature for scanning through data. If you right click within a higher-dimension image, such as the Q-E slice, then you can select the lower-dimensional plot, in this case a cut. You specify a direction and thickness and you get a new plot

window with that data displayed. By dragging the integration region in the original window you automatically update the new plot.

4. Comparison to the harmonic oscillator

To make more quantitative comparisons to the harmonic oscillator model, one can use the saved ASCII files to do plotting and fitting to the functions we have discussed. You are free to download data and use your favorite package, but it is possible to do it in MantidPlot as well.

In your MantidPlot window, read in the Q dependent data that you saved from before by using the LoadAscii algorithm. It should default to reasonable settings once the file is given, and either chose a suitable workspace name or use the one it selects. Note that you can select the units of the data, typically either Energy or Wavevector as appropriate. As before, you can plot the data by right clicking the workspace and selecting Plot Spectrum with Errors…

MantidPlot can incorporate user defined fitting functions by reading Python scripts. Use the View-Script Window menu items to bring up a python window for scripting. From the File menu, open /SNS/ARCS/IPTS-2014/shared/QHO_Q_function.py. Check that the defined function will calculate the appropriate Q dependence of the scattering for all orders (including the elastic line N=0). The Debye-Waller factor W reflects the root-mean-square displacement of the scattering atoms, in this case the hydrogen.

Execute the entire function using the python window menu, Execute-Execute All, which will incorporate the fitting function into your MantidPlot session.

Back in the main window, plot the elastic cut you made (workspace cut0) and select the plot window. Change to the fitting mode using the button with a peak and red vertical lines. The two blue vertical lines that appear in the graph indicate the fitting range and may be dragged to change what data is used. Right click within the window and click on Add other function…, then select the QHO_Q_function from the pull-down list. Click on the plus by the function name in the Fit Function window to see and set the parameters. Note that the Order should be set at the appropriate one for the data being fit, and it should be fixed by right clicking on it. It is also advisable to fit the amplitude alone first, since for this version of the function it changes dramatically for the different orders. Fitting is initiated by the pulldown Fit menu or by right clicking in the plot window.

Continue plotting and fitting the different orders of the quantum harmonic oscillator data. You may compare parameters values by displaying (double clicking) saved Table workspaces labeled as cut0_Parameters, for example. Examine your fits to see how the predictions are working.

<u>Questions</u>
How well does the Q dependence support the claim that this system consists of harmonic oscillators?
What Debye-Waller factor is found? Can you relate that to the position of a particle in a quantum harmonic well?

Hint: The answer should be W ~ 0.014 $Å^2$, corresponding to a RMS displacement of H atoms of ~0.12Å. See the short notes from former ARCS post-doc Matt Lucas about calculating the relationship between the excitation energy and the ground state particle motion.

Re-examine the elastic (N=0) and higher order inelastic data.

<u>Questions</u>
Are there features that might be biasing the fit? What could they be? How would you fix the problem?
Are there other systematic errors that might come into it?
What would be some next experiments that might point out the non-harmonic nature of this problem?

5. And beyond…

If there is time the team may decide what other experiments to try. These could include

- Measure an empty can background to improve data quality
- Try different incident energies to get better resolution or see more orders of the excitations
- Try an entirely different system, uranium nitride, to see if it is a better example of the ideal quantum harmonic oscillator