Single Crystal Neutron Diffraction in Diamond Anvil Cells: DKDP as a Test Case

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Keywords: Neutron diffraction, Diamond anvil cell, Single Crystal.

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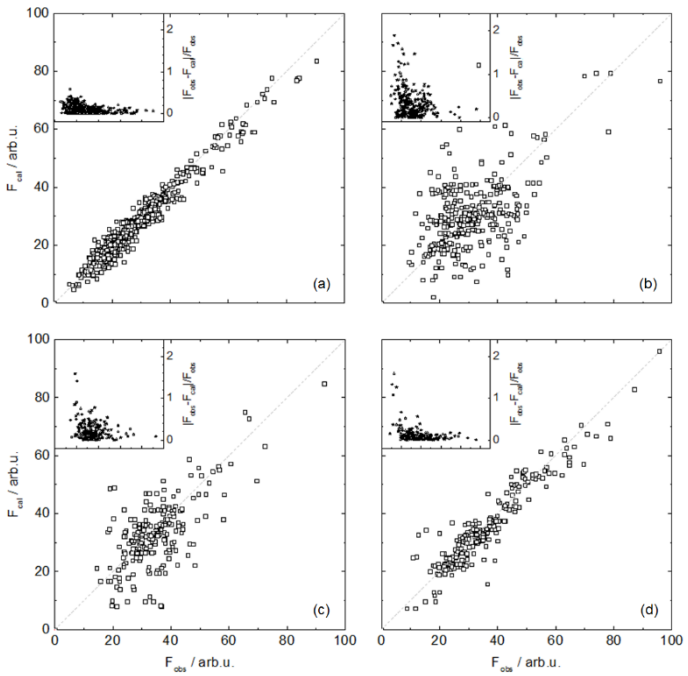
In hydrogen bonded systems, a proton is bound to one oxygen atom by a strong covalent bond and a second oxygen atom by a weaker hydrogen bond. Studies of such a bond are fascinating as they permit exploration of the behavior in response to changing relative strengths of two competing bonds. Such a system can be used to test the physics of a proton trapped within a potential well and information gleaned from such studies can have broad-ranging impact spanning diverse problems including explaining bio-chemical processes such as protein folding [1], the behavior of superionic ice in Uranus and Neptune [2,3] and quantum nuclear effects in ferroelectrics [4].

An extremely potent experimental approach to studying such phenomena is neutron diffraction. However, until recently, the measurement of neutron powder patterns of sufficient quality for structural refinement has been limited to pressures below 25 GPa. Recent developments in diamond synthesis paved the way for new large volume diamond cells. Combined with extremely bright neutron sources (e.g. SNS) neutron diffraction up to a pressure of 75 GPa has been made possible. This technique has - so far - mainly been used for powder diffraction, and as such has its limits [5].



For many systems, single-crystal techniques are critical in that they provide a much higher real-space resolution enabling access to short d-space Bragg reflections that would be heavily overlapped in a powder study. Of equal importance, they are far-better suited to diffraction studies of materials with a high hydrogen content (up to pure hydrogen), with the concentrated signal from sharp single-crystal Bragg spots remaining visible on top of potentially overwhelming incoherent scattering.

However, quantitative single-crystal neutron diffraction (SCND) studies under high pressure remain challenging, and the full possibilities of the approach are almost completely untested. Here we will show preliminary results of a trial experiment carried out on the SNAP and TOPAZ instruments at ORNL (Oak Ridge, TN, USA) whereby we will investigate the relationship between sample volume and data quality. These properties are inversely related to one another yet, unavoidably, the former is strictly limited in high-pressure studies. The systematic study of the crystallographic limitations of progressively smaller sample volumes is a critical first step in planning the development of high-pressure SCND. Furthermore, we will show the influence of the complex sample environment of diamonds and gasket on the data quality. To that end we will examine structural refinements of the deuterated ferroelectric potassium di-hydrogen phosphate (DKDP), as a test case. DKDP is a classic example of a short hydrogen bonded system, and one which has already been heavily studied – its structural properties are therefore well known.

Acknowledgments: We would like to thank the beamline scientists and technical staff of the SNAP and TOPAZ instruments for assistance with the experiments and ORNL for access to the SNS through the allocation of beamtime.

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Figure 1: Observed versus calculated structure factors as a measure of the data quality for (a) the DKDP crystal on a kapton pin and (b-c) inside a diamond anvil cell for various analysis procedures.