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Commission on High Pressure

TOWARD THE DENSER, HOTTER, FASTER AND BRIGHTER FUTURE: CHALLENGES AND OPPORTUNITIES OF HIGH PRESSURE CRYSTALLOGRAPHY

Kamil F. Dziubek[1,2], Luhong Wang[3], Haozhe Liu[3, 4]

[1]LENS - European Laboratory for Non-Linear Spectroscopy, Via Nello Carrara 1, 50019 Sesto Fiorentino, Italy
[2]Istituto di Chimica dei Composti OrganoMetallici ICCOM-CNR, Via Madonna del Piano 10, 50019 Sesto Fiorentino, Italy
[3]Harbin Institute of Technology, Harbin,150080 China
[4]HPSTAR - Center for High Pressure Science & Technology Advanced Research, Beijing, 100094, China
dziubek@lens.unifi.it, luhong1@hit.edu.cn, haozhe.liu@hpstar.ac.cn

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Abstract

Reflection upon the past makes us not only wonder how the ideas of high pressure research sprang from the minds of creative visionaries and have been evolving over the past decades, but also to think about the direction the discipline is heading today. Like in most experimental sciences, this movement is shaped predominantly by emerging techniques and instrumentation. Coupling diamond anvil cells with laser heating made it possible to generate extreme pressures and temperatures in static experiments, effectively mimicking the thermodynamic conditions of planetary interiors. Double-stage and toroidal diamond anvils pushed the limits to about one terapascal (TPa) - ten million times the atmospheric pressure. Even higher, unimaginable pressures of several TPa are achieved in dynamic compression studies, which have paved the way to a better understanding of the physics and chemistry in the depths of giant exoplanets, and also allowed us to probe fundamental properties of matter at extremes, often extending our comprehension beyond textbook knowledge. On the other hand, extremely short and bright pulses from X-ray free-electron lasers are capable of generating and investigating formerly unobserved states of matter, such as the so-called warm dense matter, representing the missing link between solid and plasma. The scientific impact of the recent cutting-edge developments on selected topics in the field of Earth's science and physics will be briefly mentioned in this report. The IUCr Commission on High Pressure (CHP) is engaged in a wide variety of activities directed at the community, including organizing annual workshops with a training session for early-stage researchers. The workshops have successfully introduced noteworthy new topics and innovative approaches, which will lead to a brighter future in this field.

The earliest *in situ* high-pressure diffraction studies date back to 1930s [1], but the major breakthrough came in 1958 with the advent of a diamond anvil cell (DAC) [2]. This small but mighty device revolutionized the world of extreme conditions research, not only boosting the achievable pressure limit but also permitting a number of experimental spectroscopic and diffraction techniques to effectively probe the sample. Soon the DAC began to evolve from its early prototype, continually striving to push the boundaries of pressure. In 1976, Ho-Kwang Mao and Peter M. Bell from Carnegie Institution of Washington obtained pressure exceeding 100 GPa (1 Mbar) for the first time [3]. Following this major step, in 1990, a team from Cornell University led by Arthur L. Ruoff, reported a new record of static pressure of 416 GPa [4], higher than at the center of the Earth (around 360 GPa). It has been noted that the logarithm of record pressure progresses linearly with time, and concluded that before the year 2000, one should overcome the limit of 1 TPa. Are we already there?

The answer is: yes and no. No because all subsequent efforts undertaken with a conventional DAC to reach pressures much in excess of 400 GPa have been in vain; it seems to be the limiting pressure for typical anvils. But yes, we are there. Thanks to the development of the double-stage DAC the magic threshold of 1 TPa has been surpassed [5], and recently toroidal diamond anvils were also shown to be possibly capable of accomplishing this task [6, 7]. However, this is not the end of the story. While static pressure experiments have their intrinsic limitations related to the mechanical durability and physical properties of diamond (*e.g.* the yield strength), this is where dynamic compression can come into play, using gas guns, laser-driven techniques or high frequency electromagnetic wave generator like Z-machines. Thus, accessible pressure range is extended to several TPa [8].

Another important factor that has recently enabled exploration into extreme condition studies is time. Time-resolved experiments are becoming more and more frequent, and undisputedly X-ray free-electron lasers (XFELs) couple naturally and fruitfully to dynamic studies, with their intense beams used either as pump or probe. For example, very short and extremely bright XFEL pulses can produce and diagnose previously unobserved states of matter, such as the warm dense matter state bridging solid and plasma [9].

It was also shown recently that dynamic compression techniques can be effectively coupled with *in situ* X-ray diffraction (XRD) at XFELs and synchrotrons. This will open new frontiers of high pressure crystallography, such as direct imaging of ultrafast lattice dynamics [10], unambiguous demonstration of the effect of rapid compression on phase boundaries [11], and *in situ* studies of phase transformation rates and pathways [12, 13].

Still, the most extreme conditions found in nature - like interiors of giant exoplanets or stars, where pressure can reach petapascals (1 PPa = 10^{15} Pa) - remain out of reach and motivate us to continue to push the boundaries of current experimental techniques. Recent important works at ultra-high pressure, such as the experimental confirmation of the insulator-metal transition in dynamically compressed dense fluid deuterium through optical measurement up to 600 GPa [14], measurement of the crystalline lattice structure and compressibility of superionic water up to 400 GPa [15], and studies of the crystal structure and equation of state of Fe-Si alloys at super-Earth core conditions over 1300 GPa [16], demonstrate that the field is ripe for new discoveries above the limits of conventional DAC experiments.

However, pressure is only one of the thermodynamic parameters characterizing the state of matter. High-pressure and high-temperature static studies were initially limited to the resistive heating of a DAC, a system not adapted to temperatures above ~ 1500 K. Only after the introduction of laser heating the temperature limit was raised to ~ 6000 K, sufficient to mimic the conditions of matter in Earth's depths. One advance made possible by hotter high-pressure environments is the creation of new stoichiometries. One such study demonstrated that the mineral goethite (FeOOH) could form FeO_2 and release H_2 under deep lower-mantle conditions, which might improve our understanding on Earth's oxygen–hydrogen cycles [17]. Another excellent case is hydrogen-bearing iron peroxide (FeO_2H_x) synthesized from the superoxidation of iron by water, which could explain the origin of ultralow-velocity zones at Earth's core–mantle boundary region [18].

In addition to many mineral physics studies, the high pressure technique is considered as powerful tool in many fields of physics. Chasing metallic hydrogen is the well-known 'Holy Grail' in high pressure physics, and a full review of the efforts since its theoretical prediction in 1935 [19] would be far beyond the scope of this short report. Although the Harvard group claimed to reach the regime of the Wigner-Huntington transition to metallic hydrogen [20], multiple groups have had different opinions on this topic [21, 22]. The crystalline structure of phase V of hydrogen under strong compression [23], still is a challenging subject from an experimental crystallographic point of view.

Strong interest in the properties of hydrides under high pressure conditions are related to another popular physics topic: high T_C superconductivity. The previous record T_C of 203 kelvin in a H-S system was achieved at high pressure conditions [24]. More exciting predictions for potential room temperature conventional superconductors in hydrogen rich compound systems [25] were experimentally confirmation very recently in La-H system with a new record T_C of 260 kelvin at 180- 200 GPa [26]. This new era for superconducting materials with higher T_C is on the way, not at ambient conditions but high pressure conditions.

This summary of the most recent achievements of high pressure crystallography, focused mainly on the experimental frontiers, is by no means exhaustive. One has yet to mention also a broad spectrum of probes, not limited only to X-ray diffraction or neutron scattering, but also spectroscopic methods, transport property measurements, etc. There are also other emerging techniques like high pressure studies in high magnetic fields. Theoretical predictions also play an extremely important role in many topics in the high pressure research field [16-20, 23-26], and more excellent cases can be found in recent review reports [27, 28].

It needs to be emphasized that high pressure tools complement and high pressure research spans an extremely wide variety of topics, ranging from geology and Earth science, physics and chemistry, material science and even biology. The IUCr Commission on High Pressure (CHP) is deeply aware of this diversity and makes every effort to properly represent all fields, and to provide balanced representation during the annual CHP Workshop sessions. Another ongoing activity of the CHP is aimed at defining essential high pressure data and metadata descriptors, in line with the IUCr Committee on Data recommendations.

In summary, 'high pressure crystallography' is today an umbrella term for all the different techniques, methods and points of interest represented in the broad community, with high pressure as the common denominator. Therefore it is essential for the CHP to follow up the new trends and innovations; to embrace the denser, hotter, faster and brighter future while being inclusive; and offer a platform for discussion, exchange of ideas, and creating collaborations.

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