Final Report

1. Cover Page

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Abstract

The grant supported a combination of experimental and theoretical research characterizing materials at high pressures (above 0.1-1 TPa = 1-10 million atmospheres) and modest temperatures (below 20,000-100,000 K). This is the "warm dense" (sub-nuclear) regime relevant to understanding the properties of planets, and also to characterizing the chemical bonding forces between atoms. As such, the experiments provide important validation and extensions of theoretical simulations based on quantum mechanics, and offer new insights into the nature and evolution of planets, including the thousands of recently discovered extra-solar planets. In particular, our experiments have documented that: 1) helium can separate from hydrogen at conditions existing inside Jupiter and Saturn, providing much of these planets' internal energy hence observed luminosities; 2) water ice is likely present in a superionic state with mobile protons inside Uranus and Neptune; 3) rock (oxides) can become metallic at conditions inside "super-Earths" and other large planets, thereby contributing to their magnetic fields; and 4) the "statistical atom" regime that provides the theoretical foundation for characterizing materials at planetary and astrophysical conditions is now accessible to experimental testing.

2. Objectives and New Experimental Approaches

The major objective of the project is to characterize warm dense matter at conditions existing deep inside planets, as distinct from stars, both to understand the current state as well as the evolution and origins of planets. Our approach combines state-of-the-art laser-based dynamic compression experiments with first principles (quantum mechanical) simulations, allowing us to enhance the predictive capability of theory. We develop and apply new experimental methods to document the equations of state, atomic-packing structures, and thermodynamic and transport properties of warm dense matter, including H_2-He mixtures, H_2O and SiO_2. By using a combination of shocks, multi-shocks and ramp compression on pre-compressed samples, we explore a broader range of pressure-density-temperature states than previously accessible. We apply unique optical and x-ray ultrafast diagnostics that our group has developed on the Omega Laser at NLUF (U. Rochester) and JANUS at LLNL, such as imaging interferometric velocimetry, optical pyrometry, broadband reflectivity and powder x-ray diffraction. Overall, this work provides an important experimental benchmark for condensed matter and HEDLP theories, as well as for planetary modeling.

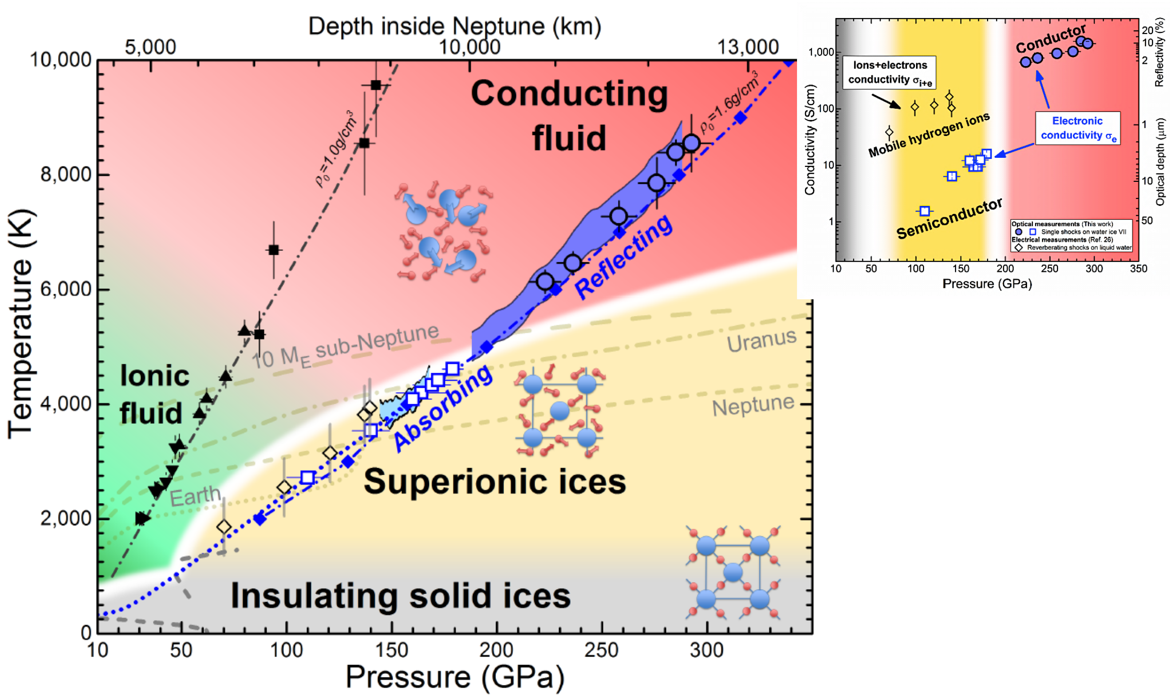

Fig. 1. Phase diagram of H_2O derived from new shock experiments on samples pre-compressed to form Ice VII (*colored symbols* indicating optically reflecting and absorbing properties at 532 nm wavelength: initial density of 1.6 g/cm^3) combined with prior Hugoniot data on liquid water (*black symbols*: initial density of 1.0 g/cm^3). The electrical conductivity calculated from our optical measurements (*inset*: upper right) are consistent with prior electrical conductivity measurements below 150 GPa and the occurrence of a superionic state of H_2O. Our new data imply that crystalline, superionic H_2O ice may be present in significant quantities inside Uranus and Neptune [Millot, et al., 2017].

3. Experimental Results

H_2O. Our work on H_2O confirms a longstanding theoretical prediction that this compound forms a superionic phase, with mobile protons moving through a crystalline lattice of oxygen, at the high pressures and temperatures existing inside such planets as Uranus and Neptune (Fig. 1). Superionic salts are of great interest in electrochemistry, including important applications to battery technologies, and our work provides a significant validation of modern first-principles calculations. We find that superionic water is likely to be far more important for understanding "icy" giant planets than previously realized. A large fraction of these planets – including many extra-solar planets – apparently consists of this crystalline state of matter, which thus controls their long-term evolution.

Our results raise for the first time the possibility that planetary magnetic fields could be produced by dynamo action in plastically deforming solid H_2O. Magnetic fields provide unique information about the internal dynamics of planets, including for extra-solar planets: aspects of the fields are in principle observable from Earth. Our finding provides evidence for an entirely new and previously unanticipated class of ("solid-state") dynamos and is therefore a significant breakthrough for planetary science, and may also offer important insights into Uranus' strange field structure.

SiO_2. Another major contribution to understanding planetary interiors came from our work on SiO_2, in which we used the high-density stishovite phase – synthesized at high pressures – rather than pre-compression in order to increase the final density (decrease temperature) of dynamically compressed samples [Millot, et al., 2015]. In this case, we showed that the rocky (silicate) portion of large terrestrial planets can contribute to core dynamos, and we found evidence that deep magma oceans may control the thermal evolution of such "super-Earths" far more extensively than is thought to have been the case for our planet (magma ocean created by the Moon-forming impact).

H_2-He. We have also completed a major study on hydrogen-helium mixtures, obtaining the first experimental evidence for fluid-phase separation in this system at high pressures and temperatures (Fig. 2). This is a long-studied topic among theoreticians, but with no experimental validation to date. Indeed, there are numerous conflicting theoretical predictions about the plausibility and nature of hydrogen-helium phase separation, with heavy He raining through the hydrogen-rich interiors of giant planets. However, there is also broad consensus of the importance of this process for understanding planetary evolution, because it provides a mechanism for the evolution of the planetary interior structure that also serves as an important source of internal heating due to gravitational energy release. The excess luminosity of Saturn, for example, is thought to be attributable to such gravitational energy release caused by helium rainout. Moreover, the phase separation reflects an important distinction in the chemical bonding in fluid hydrogen and helium at high pressures and temperatures, analogous to the immiscibility of oil and water at ambient conditions, thereby offering new key insights into chemical properties of matter at extreme conditions.

Publication of our results on H_2-He mixtures is awaiting completion of a crucial reanalysis of calibration experiments, including collecting new data on SiO_2 that serves as a key standard (Fig. 3). Agreement between data collected on different platforms (e.g., gas guns vs. Z-facility vs. laser-compression experiments) now show unprecedented levels of agreement, thus providing confidence that the 1-10 TPa regime of pressures – with forces comparable to the quantum-mechanical forces that determine atomic structure – is now accessible to reliable experimental quantification.

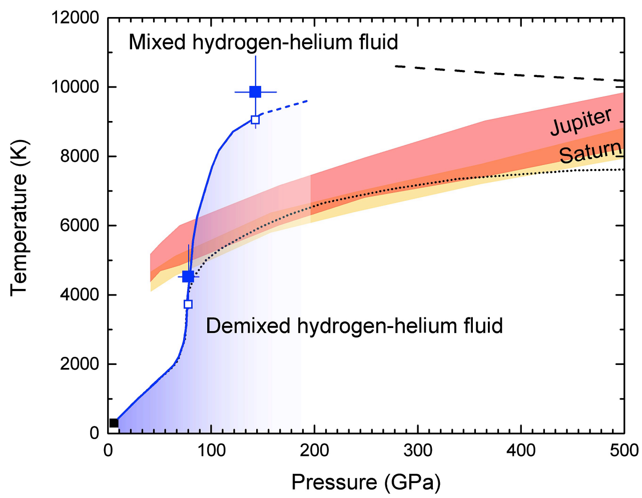

Fig. 2. Laser-driven compression experiments on pre-compressed hydrogen-helium mixtures provide evidence of phase separation below 8000-10000 K at pressures of 100-500 GPa, relevant to the interiors of Jupiter, Saturn and extra-solar giant planets [Brygoo, et al., 2017].

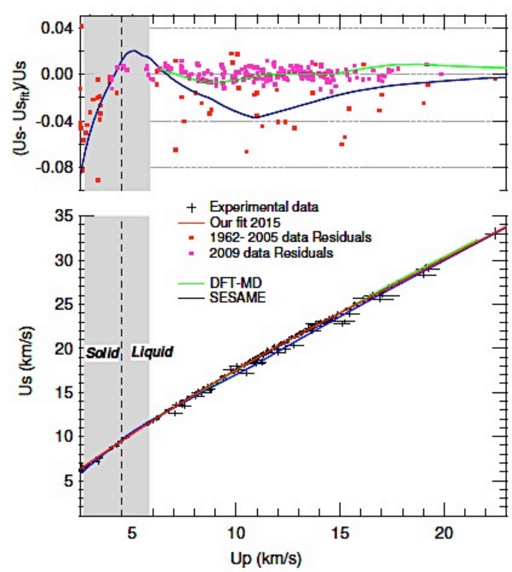

Fig. 3. New results and analysis on the shock-wave equation of state of SiO_2, a key experimental standard for laser-driven compression experiments (especially those involving pre-compressed samples). The new analysis greatly improves the reliability and precision of current measurements [Brygoo, et al., 2015].

4. Other Novel Experimental Techniques in Development

Our research program includes fielding experiments at complementary facilities (ranging from JANUS to OMEGA and NIF), and developing new methods. For example, we have developed a broad-band reflectance diagnostic that provides both wavelength- and time-resolved data on laser-shocked samples (Fig. 4). We are also developing a 3rd-generation diamond-anvil cell for pre-compression experiments at NIF and other facilities, the objective being to achieve much higher initial pressures (hence densities) than possible to date.

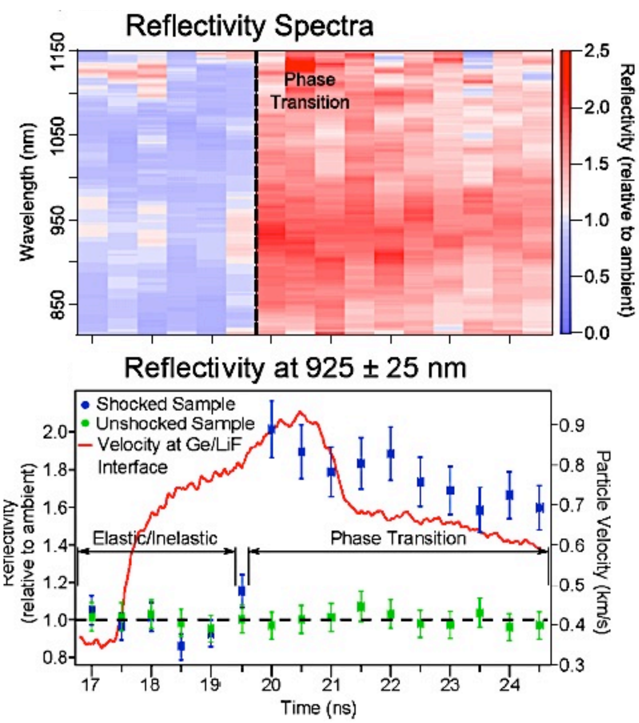

Fig. 4. Time- and wavelength-resolved reflectivity of Ge at near-infrared wavelengths under laser-driven shock loading, showing the effect of structural phase transformation on the optical properties of the sample (lower figure is a lineout at 925 nm from the upper figure) [Ali, et al, 2015].

In addition to these primary products, the project has provided partial support for a number of important complementary efforts, including validation and application of first-principles quantum mechanical calculations of the stability and properties of materials. A unique collaboration led to the first experimental characterization of carbon into the high-density regime characterized by Thomas-Fermi-Dirac (TFD) theory (Fig. 5), for instance, showing quantitative agreement between measurements and theory [Smith, et al., 2014]. This is important because the “statistical atom” approach of TFD has been the basis of astrophysical and planetary simulations for several decades, yet experimental validation has been elusive until now. We succeeded because we had access to the high energies of the National Ignition Facility (NIF), along with considerable experience with tuning the input laser intensity as a function of time to achieve shock-free ramp compression.

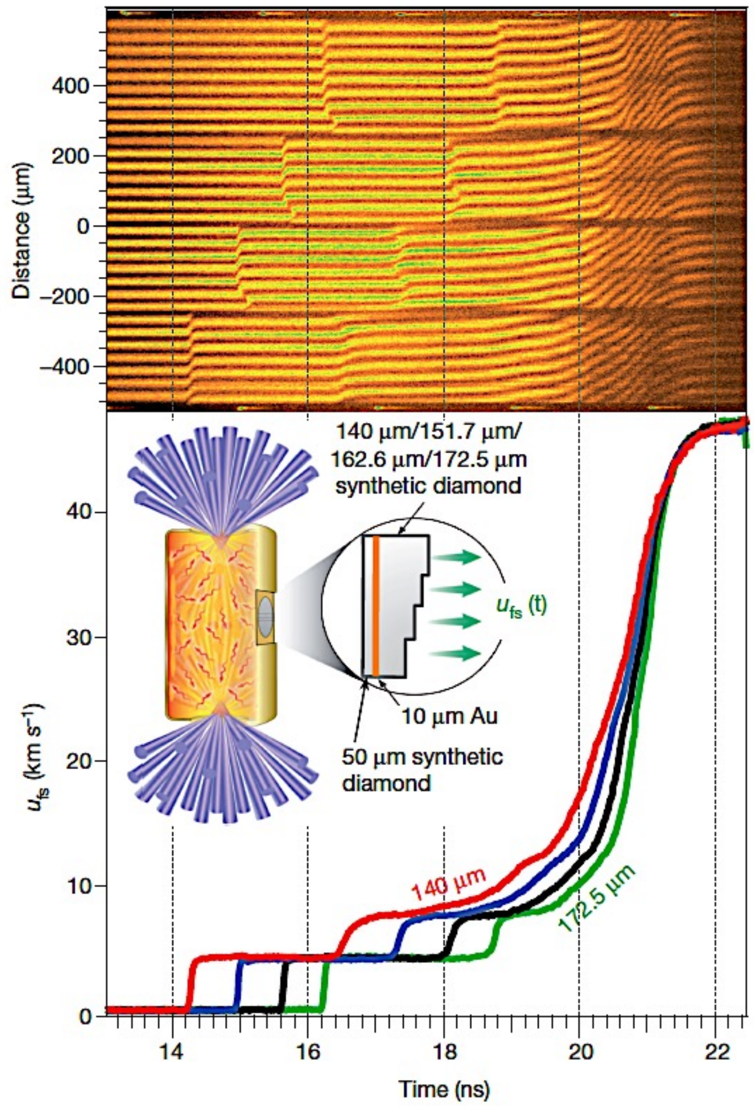

Fig. 5. Velocity interferometry record (*top*) and corresponding velocity record (*bottom*) for a diamond sample ultimately compressed to 5 TPa (50 Mbar) at the National Ignition Facility. A schematic of the sample assembly is shown in the inset (*center*) [Smith, et al., 2014].

We have also used first-principles theory to predict the properties of matter at deep-planetary interior conditions, notably the crystal structure and elastic properties of iron at TPa pressures relevant to super-Earth and other large planets [Godwal, et al., 2015]. Our research also includes understanding the complex electronic and bonding states achieved in compounds of iron and oxygen, basically simulating the combination of planetary mantles (oxides making up rock) and their metallic cores (iron alloys): subtle interplay of electron spins and crystal structure are fundamental to these compounds [Greenberg, et al., 2017].

Extrapolation of measurements under large compression to the regime of expansion (i.e., under tension) also reveals important constraints on the functional form of the binding-energy curve and equation of state (derivative of energy as a function of volume) of materials [Jeanloz, 2017]. This is useful for developing scaling relations for planets, and identifying systematic trends that do not depend on the specific material (i.e., planetary material) being considered.

Finally, we have developed novel means of making dynamic measurements in diamond-anvil cells, thereby allowing calorimetric measurements under static high pressures that complement the measurements of heat capacity and latent heats that we have realized under shock loading [Geballe, Collins and Jeanloz, 2017; Geballe, et al., 2017]. This is a major breakthrough in diamond-anvil capability that provides new thermodynamic characterization of small samples at high pressures and temperatures.

5. Products

Several publications have resulted from work to date on this project, with the research for 11 publications having been partly or fully supported by this grant (see references below). There have been more than 20 conference papers and invited presentations, including at American Geophysical Union and American Physical Society meetings. We have no new patents, but important advances have been made in experimental technologies. The research funded by this grant has supported undergraduate as well as graduate education at UC Berkeley, as well as collaboration with leading researchers at the following DOE Laboratories: Lawrence Livermore National Laboratory (LLNL), Laboratory for Laser Energetics (LLE: Rochester, NY) and Argonne National Laboratory.

6. Participants and other collaborating organizations

Dr. Marius Millot, a postdoctoral researcher at UC Berkeley and then LLNL, led the experimental effort, under the guidance of the PI and with support of Suzanne Ali and Matthew Diamond, PhD students in Chemistry and Geophysics (respectively) at UC Berkeley. The grant also supported undergraduate honors thesis work by Sarah Arveson, a geophysics major at UC Berkeley. Dr. Millot is now a staff member at LLNL, Ms. Ali completed her PhD thesis in 2015 and is now at LLNL, and Ms. Arveson is in the PhD program at Yale University. The work was carried out in collaboration with Dr. Gilbert Collins and his HED research group at LLNL (including Dr. Peter Celliers, Dr. Jon Eggert, Dr. Sebastien Hamel and Dr. Eric Schwegler), and benefitted from collaborations with the University of Rochester (Dr. Tom Boehly) and CEA in France (Dr. Paul Loubeyre and Dr. Stephanie Brygoo). The research has involved extensive collaboration between academics at UC Berkeley, Stanford, Princeton and Carnegie Institution of Washington along with colleagues at LLNL and NLUF.

7. Impact

The technique developments for pre-compressed laser-shock experiments, including diffraction, high-resolution velocity interferometry imaging and time-resolved reflectance spectroscopy, offer unique capabilities for HED experimental research, as do the dynamic-heating techniques developed for calorimetry with diamond-anvil cells. The work is targeting high-impact science in condensed-matter and chemical physics, as well as in planetary science. It has successfully supported the development and mentoring of young scientists, including toward providing candidates for positions in the National Laboratories and toward enhancing diversity in the research community. No more than $2000-5000 of the budget is estimated to have beendirected to support in foreign countries (note that even this amount serves to provide our group access to facilities in Europe and Asia from which we benefit).

References

R. F. Smith, J. H. Eggert, R. Jeanloz, T. S. Duffy, D. G. Braun, J. R. Patterson, R. E. Rudd, J. Biener, A. Lazicki, A. V. Hamza, J. Wang, T. Braun, L. X. Benedict, P. M. Celliers and G. W. Collins, Ramp compression of diamond to 5 TPa: Experiments taking carbon to the Thomas-Fermi-Dirac regime, *Nature*, **511**, 330-333 (2014).

M. Millot, N. Dubrovinskaia, A. Cernok, S. Blaha, L. Dubrovinsky, D. G. Braun, P. M. Celliers, G. W. Collins, J. H. Eggert and R. Jeanloz, Shock compression of stishovite and melting of silica at planetary interior conditions, *Science*, **347**, 418-420 (2015).

S. J. Ali, C. A. Bolme, G. W. Collins and R. Jeanloz, Development of a broadband reflectivity diagnostic for laser driven shock compression experiments, *Review of Scientific Instruments*, **86**, 043112 (2015).

S. Brygoo, M. Millot, P. Loubeyre, A. E. Lazicki, S. Hammel, T. Qi, P. M. Celliers, F. Coppari, J. H. Eggert, D. E. Fratanduono, D. G. Hicks, J. R. Rygg, R. F. Smith, D. C. Swift, G. W. Collins and R. Jeanloz, Analysis of laser shock experiments on precompressed samples using a quartz reference and application to warm dense hydrogen and helium, *Journal of Applied Physics*, **118**, 195901 (2015).

B. K. Godwal, F. Gonzalez-Cataldo, A. K. Verma, L. Stixrude and R. Jeanloz, Stability of iron crystal structures at 0.3-1.5 TPa, *Earth and Planetary Science Letters*, **409**, 299-306 (2015).

Z. M. Geballe, G. W. Collins and R. Jeanloz, Modulation calorimetry in diamond anvil cells I: Heat flow models, *Journal of Applied Physics*, **121**, 145902 (2017).

Z. M. Geballe, V. V. Struzhkin, A. Townley and R. Jeanloz, Modulation calorimetry in diamond anvil cells II: Joule-heating designs and prototypes, *Journal of Applied Physics*, **121**, 145903 (2017).

R. Jeanloz, Shock-wave and finite-strain equations of state at large expansion, in *Advances in the Computational Sciences, Symposium in Honor of Dr. Berni Alder's 90th Birthday* (E. Schwegler, B. M. Rubenstein and S. B. Libby, eds.), World Scientific, Singapore, pp. 148-161 (2017).

S. Brygoo, P. Loubeyre, M. Millot, J. R. Rygg, P. M. Celliers, J. Eggert, T. R. Boehly, R. Jeanloz and G. W. Collins, Experimental evidence for phase separation in hydrogen-helium mixtures at Jovian planetary conditions, *in review* (2017).

E. Greenberg, I. Leonov, S. Layek, Z. Konopkova, M. M. Pasternak, L. Dubrovinsky, R. Jeanloz, I. A. Abrikosov and G. Kh. Rozenberg, Theoretical and experimental evidence of a site-selective Mott transition in Fe_2O_3 under pressure, *in review* (2017).

M. Millot, S. Hammel, J. R. Rygg, P. M. Celliers, G. W. Collins, F. Coppari, D. E. Fratanduono, R. Jeanloz, D. C. Swift and J. H. Eggert, Experimental evidence for superionic water using shock compression, *in review* (2017).