Single-Shot Raman Spectroscopy Diagnostic Development for Dynamic Compression Experiments on OMEGA

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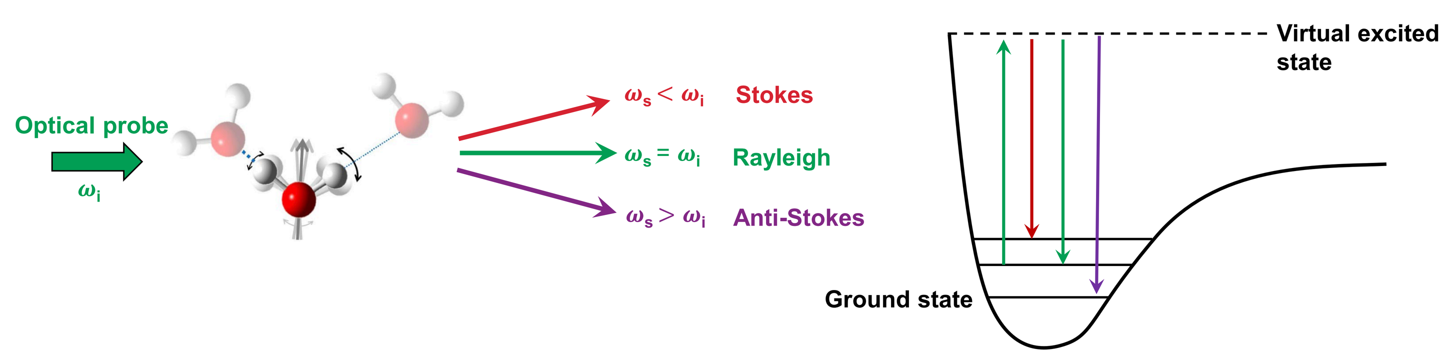
Abstract

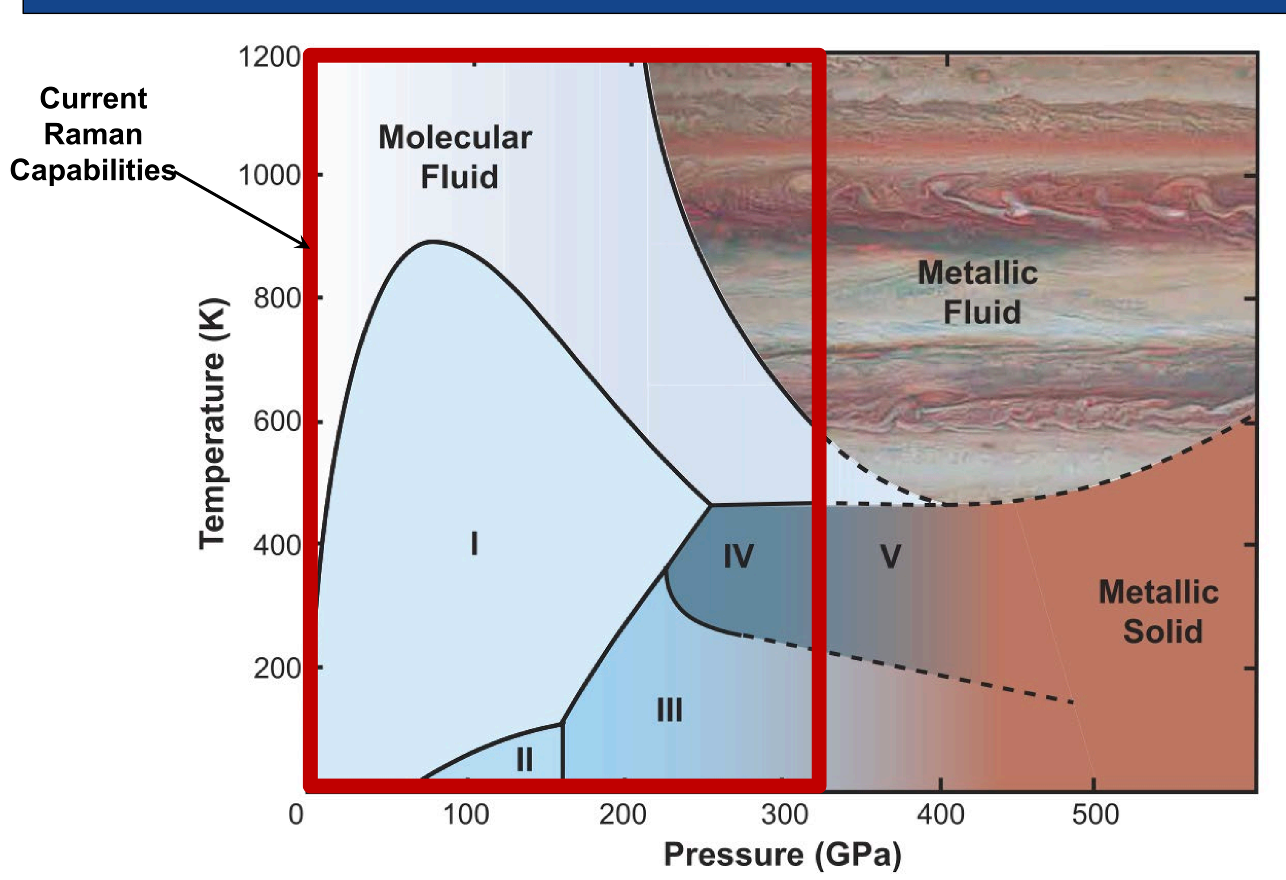
The pressures reached in dynamic compression experiments are sufficient to disrupt the electronic structure of matter, often inducing chemical and physical transitions. These dynamic processes are not directly measured by existing diagnostics that rely on a material's ability to scatter x rays. To this end, our work will develop a time-resolved Raman spectrometer for the OMEGA Laser System at the Laboratory for Laser Energetics. This diagnostic will measure the intensity and wavelength of a Raman-backscattered 395-nm probe from laser-compressed materials. This diagnostic will directly probe the evolution of bond character of materials at high-energy-density conditions

Motivation

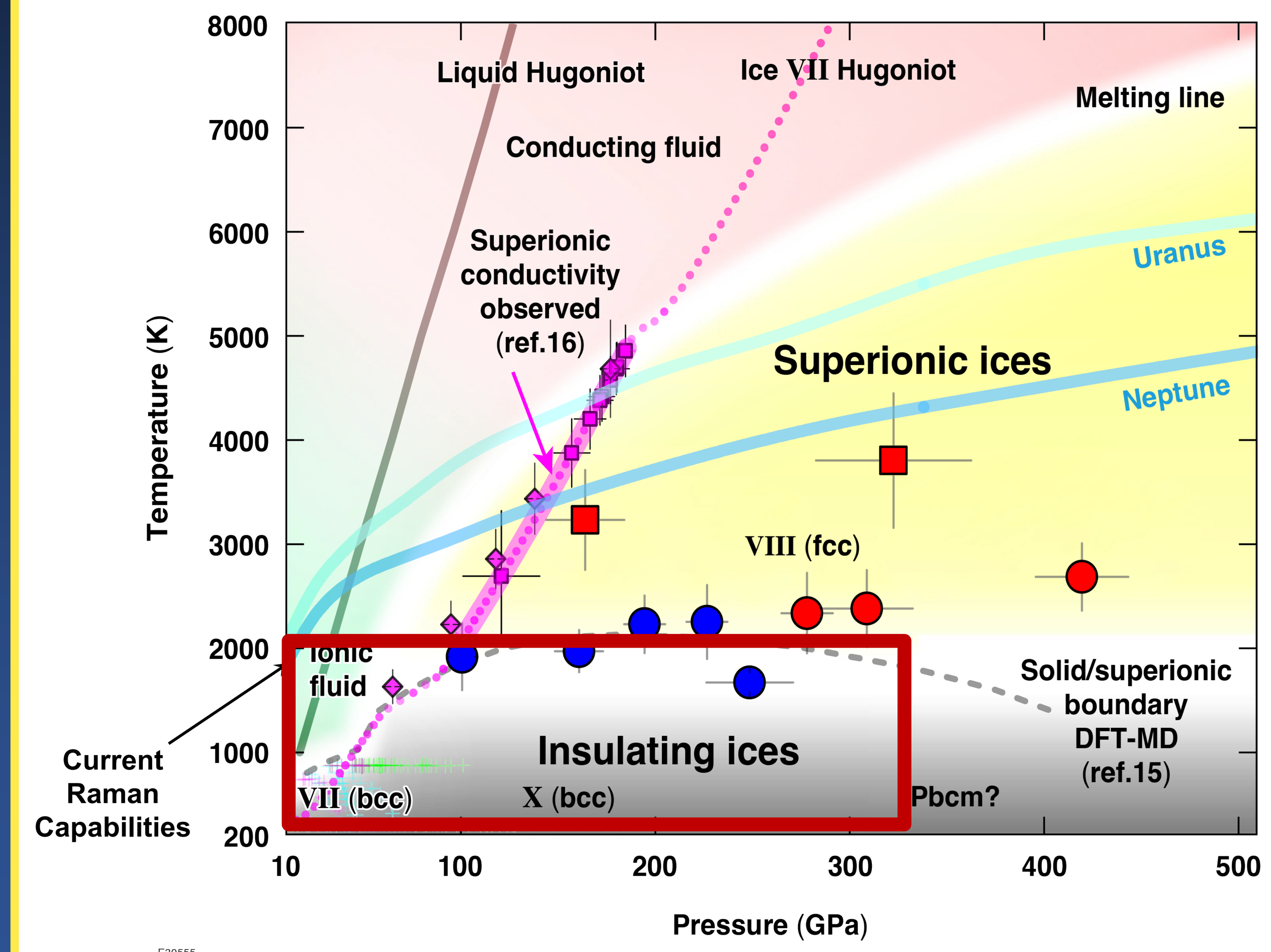
There is currently no way to directly measure chemical bonds in matter at high energy density science condition. Raman spectroscopy is a technique that directly measures elementary excitations and is highly sensitive to symmetry and bond character. Therefore, single-shot, nanosecond-scale, Raman spectroscopy at OMEGA will explore, for the first time, dynamic changes in chemical bonding in high energy density matter



(Above) Incident photons a sample excite a sample to a virtual excited state.[1] These photons can be inelastically scattered at a different frequency based on the sample's final elementary excitation state (e.g., rotational, vibrational)

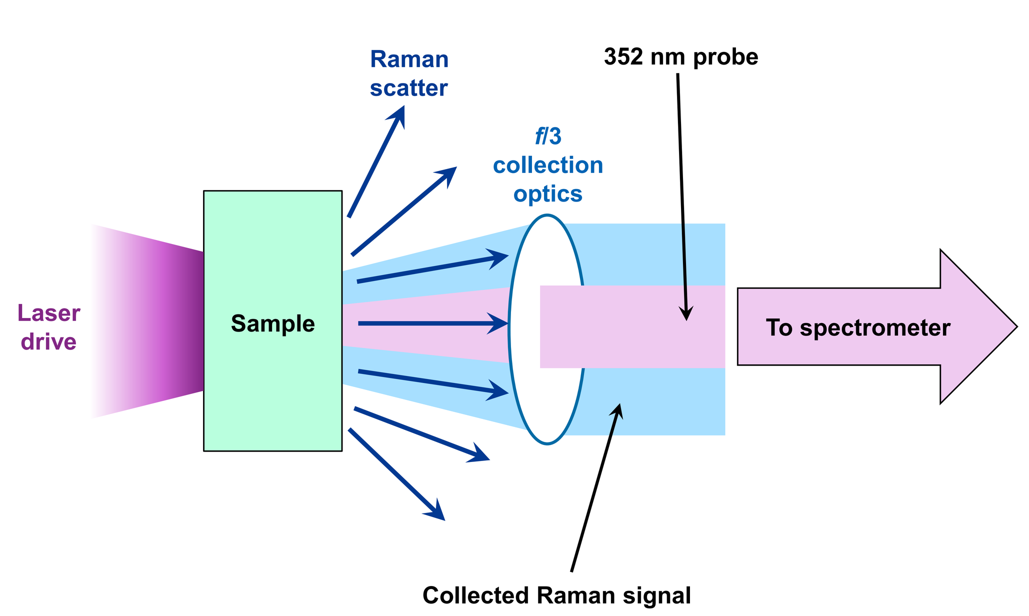


(Above) Phase diagram of hydrogen showing the predicted region of solid metallic hydrogen.[2] Raman spectroscopy for dynamic compression experiments will be used to investigate the structure of solid hydrogen above this pressure.



(Above) Phase diagram of water.[3] Raman spectroscopy will be used during dynamic compression experiments on water to measure the dissociation of hydrogen from oxygen in the onset of superionicity.

Experiment

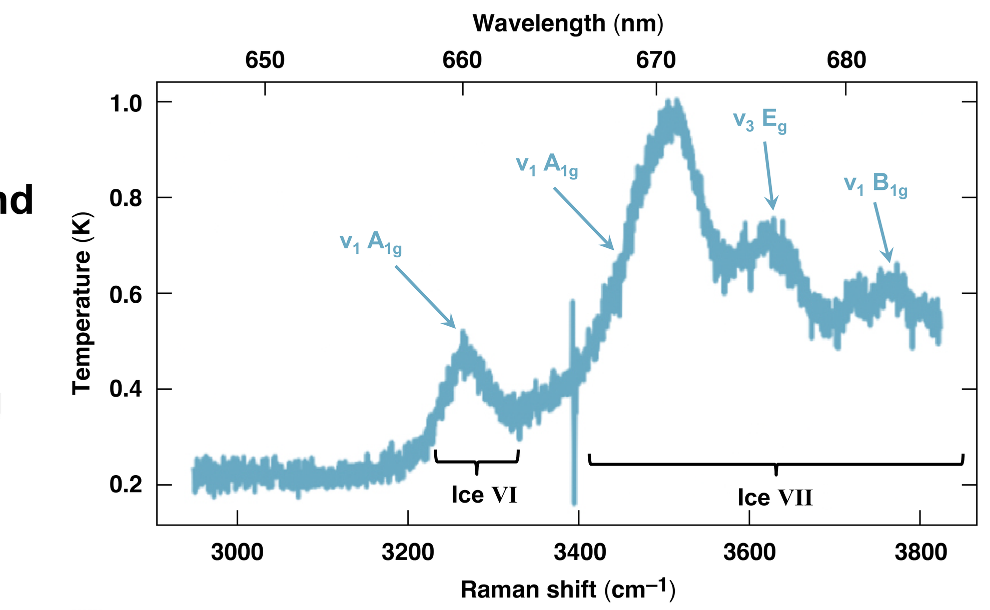


(Left) Diagnostic set up will use a backscattering geometry to bring in an external probe laser and extract Raman signal from the same port.

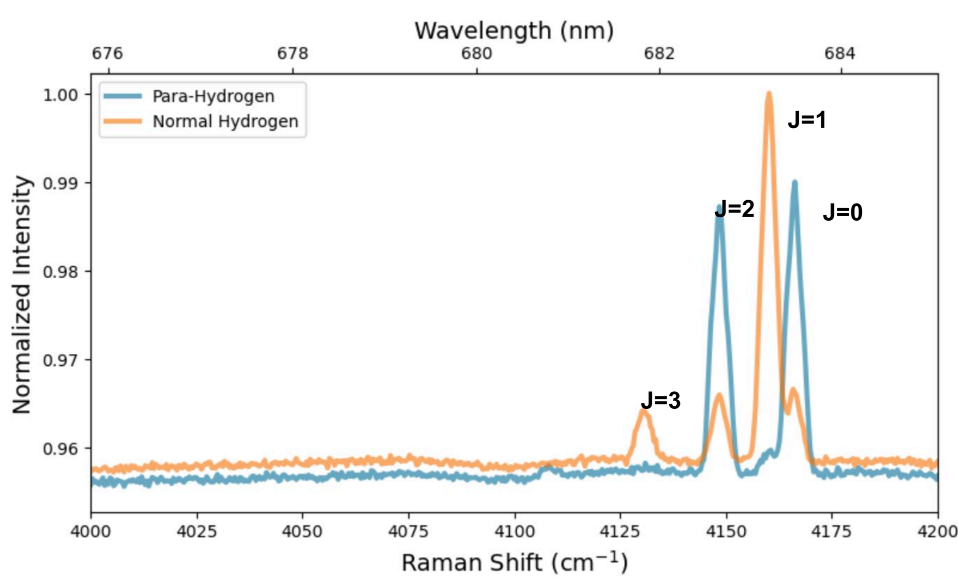
Experimental Challenges	Solutions
Performing Raman on a nanosecond scale runs the risk of low signal	• Maximize intensity of the probe laser without perturbing the sample
Experimental design requires optics that bring in an external probe and collects signal from the same port	• Make use of infrastructure that already exists on OMEGA
Raman signal will be competing with large background from thermal emission	• By using a blue probe laser, we shift the Raman signal out of the wavelength regime where most thermal emission is occurring

Preliminary Results

(Left) Sample Raman spectra of water compressed in a diamond anvil cell (DAC) to approximately 2 GPa at room temperature. The spectra was taken using a 543 nm 5 mW probe laser with a 100 ms exposure time



The initial design for the Raman diagnostic was built and tested in an offline lab. Sample spectra using equivalent input energy resulted in a spectra with a signal to noise ratio of ~20. Raman intensities are proportional to $1/\lambda^4$, so photon statistics are expected to improve by a factor of 3.6 by switching to a probe wavelength of 395 nm



(Left) Raman spectroscopy set up was used in our offline lab to determine the success of para-hydrogen procedure before dynamic compression.

Future Direction

We intend to test the viability of a Raman spectroscopy diagnostic using existing infrastructure for the two-dimensional imaging velocimeter and an optical set-up for offline testing. Iterations on probe intensity and quality will require the addition of beam conditioning optics. After successful implementation of a nano-second scale Raman spectroscopy diagnostic, we intend to add time-resolution to the diagnostic capability through the use of a fast-gated Hybrid Complimentary Metal Oxide Semiconductor (hCMOS) imaging

References

[1] Yu, CC., Chiang, KY., Okuno, M. et al. . Nat Commun, 11, 5977 (2020).
[2] Eugene Gregoryanz, Cheng Ji, Philip Dalladay-Simpson, et al. Matter Radiat. Extremes 5, 038101 (2020).
[3] Marius Millot, et. al. Nature, 569, 251 (2019).