White Paper for *Frontiers of Plasma Science Panel*

Date of Submission:	***06/19/15***

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	"P", "S"
• Plasma Atomic physics and the interface with chemistry and biology	S
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• Plasma self-organization	
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Indicate type of presentation desired at Town Hall Meeting.

	"X"
Oral	
Poster	
Either Oral or Poster	
Will not attend	X

Title:	Measurement of de-mixing and phase separation of high energy density mixtures
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- ***Describe the research frontier*** **and importance of the scientific challenge.**

High-pressure mixtures of multiple atomic species govern the internal structure and evolution of many astrophysical objects, such as giant planets, brown dwarfs or the outer crust of neutron stars and white dwarfs [1,2]. Here, gravity forms an environment for the lightest elements to be compressed to densities of several grams per cubic centimeter and heated to temperatures in the order of chemical bonding and above. Matter with such parameters is in a partially ionized, fluid state with a well-pronounced short-range order within the ion subsystem. In this so-called "warm dense matter" regime, neither lattice symmetries as in solids nor the ideal mixing as in ideal plasmas can be employed.

De-mixing and the subsequent segregation of different elements in this regime can play a crucial role in the energy balance of planets [3]. Moreover, the predicted separation into helium-rich and helium-poor phases determines a layer boundary in gas planets like Jupiter and Saturn, which has far-reaching implications on our understanding of planetary formation. Phase separation in hydrogen-helium mixtures has been observed in quantum simulations for conditions found in the interior of gas giants [4,5]. However, real experimental tests of these models within astrophysical settings can only be realized indirectly, e.g. by studying the gravitational and magnetic field structures of these planets as well as the surface temperature. Thus, interpretation of such measurements requires full scale modeling of planets. Therefore, we propose laboratory experiments to investigate these possible de-mixing and phase separation effects in warm dense mixtures.

Furthermore, the characteristics of such mixtures, on much shorter time scales, are highly relevant for particular applications such as inertial confinement fusion. Here, controlling the mix of ablator material into the fuel has been shown as one of the key issues in order to achieve high quality implosions. In addition, the so far most commonly used ablator material is plastic, which itself is a mixture of carbon and hydrogen. Thus, phase separation of carbon and hydrogen is a potential concern for this ablator material and this phenomenon could lead to local density fluctuations, as local carbon and hydrogen clusters are forming. In turn, this could seed hydrodynamic instabilities. Such instabilities, in particular ablation-front Rayleigh-Taylor growth, were identified as one of the primary issues that led to reduced implosion performance during the National Ignition Campaign, where the first shock of a so-called low-adiabat implosion drive creates pressures of 100-200 GPa and temperatures of 0.5 eV -1 eV in the ablator material.

Phase separation of hydrocarbons is also supposed to happen inside the icy giants of our solar system, Uranus and Neptune, where methane is found to be highly abundant in the atmospheres. In this context, popular science texts usually speak about "diamond rain" or "seas of liquid carbons". Due to de-mixing, molecular hydrogen may be released from the ice layers of both planets. This could lead to more compact cores in Uranus and Neptune, in contrast to recent predictions of core erosion for Jupiter and Saturn. Moreover, hydrogen leaving the core as well as accompanying non-metal to metal transition of carbon is supposed to trigger convection currents, which could explain the unusual magnetic fields of Uranus and Neptune [6].

Using laser-heated diamond anvil cells (DACs) and shock experiments with gas guns, onsets of these de-mixing processes were found. In the case of hydrocarbons, it was observed that the formation of carbon-carbon bonds is preferred in the 10-60 GPa pressure regimes and below the carbon melting temperature. First, this leads to the formation of polymers [7] and then, at higher pressure and temperature, the formation of separate diamond/liquid carbon and hydrogen clusters is predicted to start around 200 GPa or even lower [8], but has not been measured so far. Hereby, the separation of carbon and hydrogen is expected to be more favorable with increasing pressure and temperature [9,10], as long as remaining below the diamond melting temperature of ~8000 K. These conditions cannot be achieved in static DAC experiments and thus dynamic compression experiments are required. The time scale of these transitions is expected to be in the order of few picoseconds and thus, it is principally possible to observe de-mixing effects in nanosecond-timescale laser experiments.

- ***Describe the approach to advancing the frontier and indicate if new research tools or capabilities are required.***

Laser-driven single-shock, double-shock and ramp compression allow for creating conditions, which can be found deep inside giant planets. Considering hydrocarbons, corresponding samples will first pass the regime where polymers or even nano-diamonds are formed before approaching the carbon melting line and/or a transition to a plasma state.

The microscopic ion structure and thus the phase separation can be probed by spectrally resolved X-ray scattering [11]. So far, there does not exist any comparable method for direct measurement of the ion correlations at these conditions. The scattered radiation power per solid angle is given by [12]

$$\frac{dP}{d\Omega} = I_0 r_0^2 \frac{1}{2} N\left[1+\cos(\theta)\right] S(k),$$

where I_0 is the initial probe intensity, r_0 the classical electron radius, θ the scattering angle, and N the number of atoms in the probe volume. S(k) denotes the frequency-integrated electron structure factor,

which contains all the information of the ion structure in the sample. For non-collective scattering, i.e. scattering from single electrons, the structure factor can be decomposed into three terms [13,14]

$$S(k) = W_{el}(k) + W_{bound-free}(k) + W_{free-free}(k)$$

The first term describes elastic scattering, which is mainly dominated by scattering from tightly bound electrons. The second term accounts for the inelastic scattering from weakly bound electrons and the third term describes inelastic scattering from free electrons. The elastic scattering W_{el} is highly sensitive to the ionic structure of the system and for a mixed or de-mixed state, strong changes in the elastic scattering amplitude are expected. The weight of the elastic scattering in a multicomponent system is defined as follows [15]:

$$W_{el}(k) = \sum_{\alpha,\beta} \frac{\sqrt{n_\alpha n_\beta}}{n_i} \left[f_\alpha(k) + q_\alpha(k) \right] \left[f_\beta(k) + q_\beta(k) \right] S_{\alpha\beta}(k).$$

Here, the summation runs over different ion species, α and β, with the corresponding ion densities, n_α, n_β, respectively, n_i denoting the total ion density in the system. In this generalized version, the form factors f(k) and the screening functions q(k) associated with a specific ion species α, β are required. These quantities can be calculated with DFT for various ionic and electronic structures, where in most cases, the single atom form factors are a reasonable approximation [16]. The partial structure factors $S_{\alpha\beta}(k)$ contain all the structural information of the ionic subsystem and, thus, all mutual correlations between the different ion species. Thus, the elastic scattering amplitude is very sensitive to mixed or de-mixed states. On the other hand, the inelastic scattering strength is barely affected by these structural transformations and can be used for normalization.

For the realization of such experiments, a controlled high-quality drive such as multiple shock compression or ramp compression has to be combined with a brilliant X-ray source capable of delivering enough photons for spectrally resolved X-ray scattering. Adequate separate capabilities are nowadays available at modern high-energy laser facilities like the Laboratory of Laser Energetics or the National Ignition Facility. The difficulty lies in combining the methods, e.g. controlling the drive with a VISAR system and/or radiography while performing spectrally resolved X-ray scattering measurements within the same experiment. This requires development of new experimental and diagnostic capabilities at these facilities.

A very promising development is the combination of high-energy lasers with X-ray free electron lasers, as realized at the MEC endstation of the Linac Coherent Light Source. Here, high repetition rate scattering experiments at very small scattering angles are possible, which can be extremely sensitive to mixing/de-mixing or phase separation. However, while great results can already be obtained, better pulse shaping capabilities and more laser energy at MEC would be certainly helpful to better match the interesting conditions in the interior of giant planets in theses experiment.

- ***Describe the impact of this research on plasma science and related disciplines and any potential for societal benefit.***

The understanding and control of high-energy density mixtures is key in order to realize efficient inertial confinement fusion implosions as a possible source of relatively clean energy. Moreover, studying the chemistry of high-pressure and high-temperature mixtures in the laboratory will highly improve our understanding of the internal structure and evolution of planets and thus the formation of our and other solar systems up to creating the environment necessary for life on Earth and maybe other planets.

References (Maximum 1 page)

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Figures (maximum 1 page)

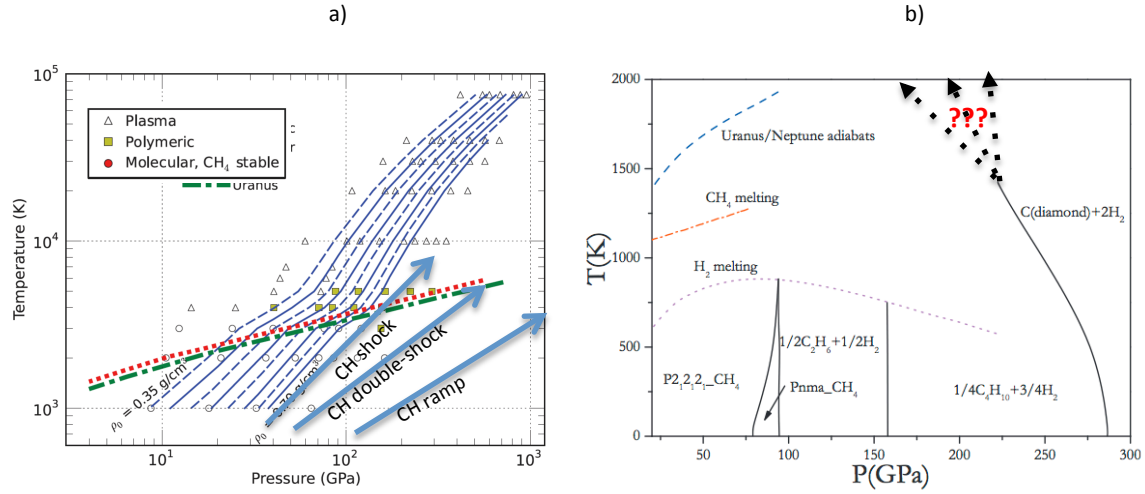

Figure 1: a) Pressure-temperature diagram of methane shock Hugoniot curves for different initial densities [17]. Single-shock double-shock and ramp compression of CH or CH_2 with initial densities around 1g/cc can reach conditions comparable to the deep interiors of ice giants like Neptune and Uranus. b) Calculation of CH separation at relatively low temperatures [18]. The separation pressure decreases with increasing temperature. The behavior towards high temperatures intersecting the Neptune/Uranus adiabats and above is subject of speculation. Thus, measurements are needed.

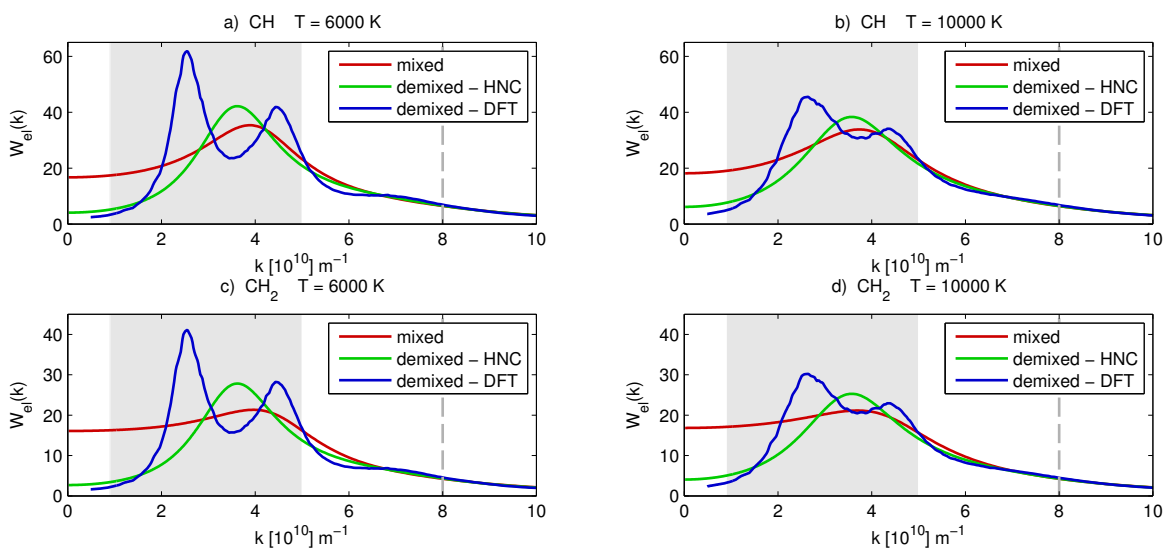

Figure 2: Simulated elastic scattering amplitudes for mixed and de-mixed hydrocarbons at different temperatures. The gray-shaded area marks the k-vector regime, which can probed by spectrally resolved X-ray scattering in order to determine the amount of de-mixing as well as validity of corresponding structure models. Large k-values as e.g. marked by the grey dashed line can be used for normalization of the elastic scattering at smaller scattering angles.