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Measurement of The Dynamic Response of Compressed Hydrogen by Inelastic X-Ray Scattering

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Abstract. Measurement of the dynamic properties of hydrogen and helium under extreme pressures is a key to understanding the physics of planetary interiors. The inelastic scattering signal from statically compressed hydrogen inside diamond anvil cells at 2.8 GPa and 6.4 GPa was measured at the Diamond Light Source synchrotron facility in the UK. The first direct measurement of the local field correction to the Coulomb interactions in degenerate plasmas was obtained from spectral shifts in the scattering data and compared to predictions by the Utsumi-Ichimaru theory for degenerate electron liquids.

1. Introduction

A central feature of planetary research and laboratory astrophysics is the creation of extreme high pressure, high temperature states of hydrogen and helium. At solid densities and high pressures (above a few GPa) degeneracy and strong inter-particle coupling start to play an important role in determining the equation of state (EOS) of the hydrogen, which is still largely unknown in this regime. To date, most experimental efforts have concentrated on creating high density and temperature conditions in fast shocks created by laser ablation [1, 2]. State-of-the-art EOS measurements of laser-driven shocks on pre-compressed H and He samples have been demonstrated using Velocity Interferometer System for Any Reflector (VISAR) diagnostics [3, 4]. While the laser driven dynamic compression reaches higher pressures, diamond anvil cells (DAC) that compress the samples statically can be used to create steady state conditions which are more representative of the planetary interiors than laser-shock experiments [5]. X-ray Thomson scattering (XRTS) [6] is capable of providing more EOS information, such as electron temperature, density and ionization state, and VISAR is less applicable at higher pressures while XRTS can still be used in such conditions, making it the preferred method for measurements of the dynamic response of highly compressed H.

2. Experimental Setup

This proof-of-principle experiment was conducted on the I15 (Extreme Conditions) beamline at Diamond Light Source, UK. The hydrogen gas loaded inside a 0.1 mm thick gasket with diameter of 250 μm was compressed using a diamond anvil cell and probed by monochromatic 19,952 eV X-rays from the synchrotron, figure 1. The inelastic X-ray scattering spectra from hydrogen DAC's at $P_1 = 2.8$ GPa (liquid phase) and $P_2 = 6.4$ GPa (solid phase) were observed in Von Hamos geometry using a curved highly annealed pyrolytic graphite (HAPG) crystal [7] coupled to a photon-counting detector (Pilatus 100k) with scattering angle of 32.2±1.1°, corresponding to a regime where the magnitude k of the scattering wavevector is comparable to the Thomas-Fermi screening wavenumber of the electrons ($k_{TF} = 1.7$ x 10^{10} m^{-1}). The DAC was positioned with 20 μm accuracy, corresponding to an uncertainty of 10 μm ($\sim$ 1 eV) at the detector plane. Uncertainties in the synchrotron energy (as determined using a Pd filter) were also found to be within 1 eV.

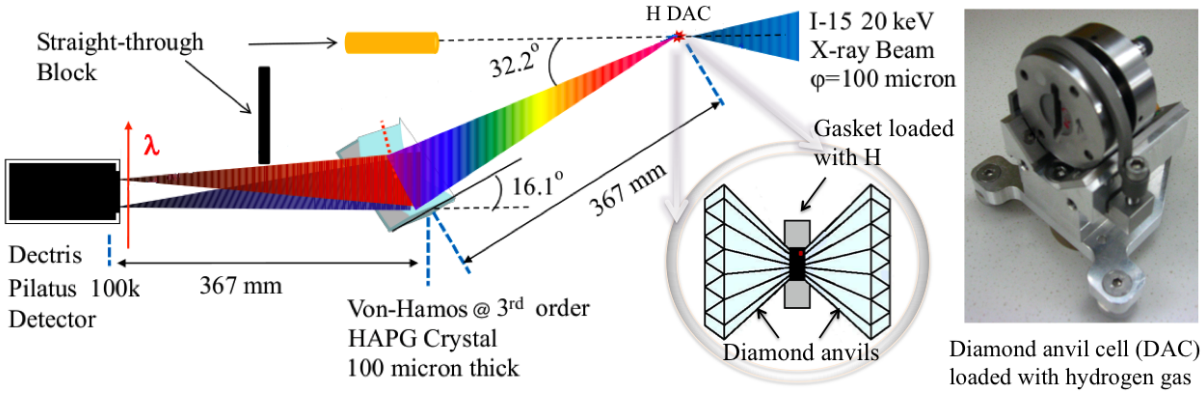

Figure 1. Apparatus arrangement.

The pressure within the anvil cell was measured through the well known relationship for fluorescence of ruby under pressure [8] (a small ruby probe was buried inside the anvil cell together with the hydrogen sample and the fluorescence was measured using an optical laser and a calibrated spectrometer). Since the pressure and temperature of the hydrogen in the anvil cell are known (room temperature, T = 300 K) the electron density could be determined from the equation of state (EOS) data for compressed hydrogen experimentally determined by ref. [5] and was found to be 6.0 x 10^{22} cm^{-3} for P_1 and 7.9 x 10^{22} cm^{-3} at P_2 at which hydrogen is fully degenerate (see table 1).

Table 1. Physical parameters in compressed hydrogen. The Wigner-Seitz radius given by $d = (3/(4\pi n_e))^{1/3}$ is the inter-particle spacing at electron density n_e and a_B is the Bohr radius.

	$P_1 = 2.8$ GPa (liquid)	$P_2 = 6.4$ GPa (solid)
Brueckner parameter, $r_s = \frac{d}{a_B}$:	2.99	2.74
Fermi energy, $E_F = \frac{\pi^2\hbar^2}{2m_e}\left(\frac{3n_e}{\pi}\right)^{2/3}$:	5.6 eV	6.7 eV
Degeneracy parameter, $\theta = \frac{k_B T}{E_F}$:	0.0046	0.0039
Plasma frequency, $\omega_p = \left(\frac{n_e e^2}{\varepsilon_0 m_e}\right)^{1/2}$:	9.12 eV	10.4 eV

3. Reduction Analysis

The weak H scattering compared to the diamond is a result of the small sample volume needed to achieve high compression of the hydrogen gas within the larger anvils and the fact that each C atom has more electrons to scatter from than the H atoms. In order to compensate for the dominant Compton scattering from the diamond anvils, we retrieved the hydrogen scattering data in the form of a ratio of spectra from hydrogen at P_1 and P_2 through a careful reduction analysis procedure using the measured signal referenced to the empty cell.

The ratio of the 2.8 GPa to 6.4 GPa spectra was computed using the following equations. The empty cell scattering spectrum corresponds to:

$$I_0 = B \cdot S_{anvil} \tag{1}$$

And the scattering spectrum of hydrogen loaded DAC at pressure P:

$$I_P = A_P \cdot (S_{anvil} + S_H^P) \tag{2}$$

Where A and B are calibration constants, which depend on the absolute response of the measuring apparatus and their ratio is a well defined quantity given by the ratio of average background noise levels of the empty and H loaded DAC spectra, and S_{anvil} and S_H^P are the dynamic structure factors from the diamond anvils only and hydrogen at pressure P, respectively:

$$\frac{S_H^{P_1}}{S_H^{P_2}} = \frac{(B/A_{P_1})(I_{P_1}/I_0) - 1}{(B/A_{P_2})(I_{P_2}/I_0) - 1} \tag{3}$$

Since the quantity on the RHS of equation (3) is directly obtained from the experimental data using the measured spectra from the empty cell and the DAC's loaded at the two pressures, the ratio $S_H^{P_1}/S_H^{P_2}$ is thus well defined.

4. Local Field Correction

Since we only obtained the ratio of the scattering signals of hydrogen at the two different pressures, the dynamic structure at a given pressure has to be retrieved from this ratio using a known result for the other pressure. Thus, in the next step, the experimentally determined ratio was multiplied by the spectrum calculated for P_2 obtaining the response at 2.8 GPa (blue line in figure 2). P_2 has been chosen, because at the higher pressure we have polycrystalline solid hydrogen (hexagonal close packed) [9]. The electrons are effectively free as they satisfy the Bloch functions for electronic motion inside the periodic potentials within the crystalline lattice of localized ions. Since, in our experiment, k lies outside the Brillouin zone boundary, the lattice does not influence scattering from the electrons. This phase is also more degenerate (lower degeneracy factor $\Theta = k_B T/E_F$) resulting from higher Fermi energy making the effects of finite temperature less important. Therefore the calculation of the scattering spectrum for P_2 can be justifiably carried out using the Random Phase Approximation (RPA) at T = 0 K [10]. The electron-electron dynamic structure factor for electrons interacting via Coulomb forces is given by the Fluctuation Dissipation Theorem, for $k_B T \ll \hbar\omega$:

$$S_H^P(k,\omega) = \frac{\varepsilon_0 \hbar k^2}{\pi e^2 n_e} \cdot \mathrm{Im}\left[\frac{-1}{\varepsilon(k,\omega)}\right] \tag{4}$$

where n_e is the electron density, e the electron charge, ε_0 the permittivity of free space, k is the magnitude of the change in wavevector due to the scattering and ω is the inelastic energy exchange. The dielectric function $\varepsilon(k,\omega)$ relates to the electron susceptibility $\chi(k,\omega)$ as $\varepsilon(k,\omega) = 1 - (e^2/\varepsilon_0 k^2) \cdot \chi(k,\omega)$ [13]. The electron susceptibility can also be written as:

$$\chi(k,\omega) = \frac{\chi_0(k,\omega)}{1 + \frac{e^2}{\varepsilon_0 k^2} G(k,\omega)\chi_0(k,\omega)} \tag{5}$$

where χ_0 is the Lindhard susceptibility at T = 0 K and $G(k,\omega)$ is the local field correction [13], which is a measure of the effects of strong coupling due to higher order interaction terms and incorporates the short-range correlations, both neglected by the RPA. For $G(k,\omega) = 0$, $\chi(k,\omega) = \chi_0(k,\omega)$ and the RPA is reproduced. While the dynamic response for liquid hydrogen is obtained from Eq. (3) using the RPA approximation for the solid hydrogen, equation (3) can also be directly applied to retrieve the liquid response if the local field is known. In case of liquid hydrogen, the theoretical value of the local field correction can be estimated using the Utsumi-Ichimaru equations in the static approximation to be $G(k,\omega) = G(k) = 0.91$ [11], and the spectrum calculated using this value of G corresponds to the dashed green line in figure 2.

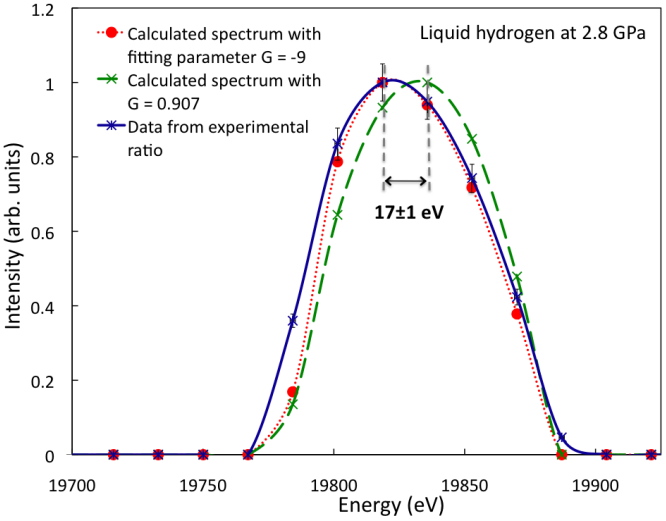

Figure 2. Normalized plots of the electron-electron feature of the scattering spectrum for H at 2.8 GPa.

There is a noticeable shift of $\sim 17 \pm 1$ eV between the peaks of the calculated spectrum using the Utsumi-Ichimaru theory and the data obtained from experiment (assuming a nearly free electron gas for the solid H), which is a result of our assumptions breaking down in the case of liquid hydrogen, where the electrons are no longer free and short range correlations can become important. The calculated response overlaps with the experimental data for $G(k = 5.59 \pm 0.19$ x 10^{10} m^{-1}, $\omega \sim 130$ eV$) = -9 \pm 1.5$ (red dots, figure 2). However, since $\omega \sim 130$ eV is significantly larger than plasma frequency ω_p, static approximations, such as the one of Utsumi-Ichimaru, may not be applicable here. Our results are the first experimental measurement of local field effects across the liquid to solid phase transition in hydrogen. They indicate a much stronger effect of correlations than suggested by fluid theories. This is important since the local field correction can be linked to dynamic properties of the compressed material, namely the isothermal compressibility and conductivity [11]. Building upon the results presented in this study, we would propose to expand and improve the XRTS method at free-electron X-ray laser facilities (FEL), where laser-driven DAC's [4] can be both heated and further compressed to the warm dense matter regime relevant to the planetary interior regime, and studied with a high-power coherent X-ray source.

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