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TEMPERATURE MEASUREMENTS AND HYDROGEN TRANSFORMATION UNDER DYNAMIC COMPRESSION UP TO 150 GPA.

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Abstract. Lithium fluoride single crystal window was used for optical light emission registration during quasi-isentropic compression of hydrogen to the pressures 100-150 GPa. Initially gaseous hydrogen samples at 78 K temperature and different pressures in the range 3-30 MPa were investigated. Recorded brightness temperature profiles in near infrared range of wavelength were analyzed to evaluate optical and transport properties of the investigated hydrogen sample and window. Two EOS models of hydrogen, with and without metallic region were used for 1-D simulation of it's properties under dynamic compression and estimation of hydrogen temperature within compressed layer. The obtained data demonstrate abrupt change of final temperatures after heating higher then 3500K.

Keywords: Hydrogen, phase diagram, equation of state, metallization.
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INTRODUCTION

Hydrogen phase diagram is not well defined in the region of temperatures 2 - 10 kK and densities of about 0.4-2 g/cc up to now [1]. According to the pioneer work of E. Wigner and H. B. Huntington [2], hydrogen at low temperatures and high pressures can be in a metal phase. There are experimental results, which show significant electrical conductivity in this region of phase diagram [3,4]. Temperature ionization can't explains registered high level of conductivity. A question, how this transition to conductive state occurs, still exists.

One of mechanism is a dramatic lowering of hydrogen ionization potential with pressure and density, which leads to formation of the region of unstable states in hydrogen [4]. According to this model, all states of liquid hydrogen can be described by a single EOS; hydrogen will be separated to coexisting metal and dielectric phases in the region of instability (two-phase region). It is not correct to separate metal and dielectric states of hydrogen in the rest of phase diagram. This transition is analogous to liquid-vapor phase transition (LV type of plasma phase transition (PPT)). Recently, such a PPT was connected with the process of hydrogen dissociation at high pressures [5].

Another mechanism, proposed in [3], assumes that metal states can exist as a separate phase with electronic structure, different from that of partially dissociated and ionized "dielectric" phase. In the metal state there will be significant electron density on the boundary of the atomic cell, contrary to the dielectric state. This prevents the mixing of phases. Thus, we have two different equations of states for metal and dielectric phases of hydrogen for any

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point in the region (temperatures 2 - 10 kK and densities of about 0.4-2 g/cc), and coexistence curve, separating metal and dielectric states. This transition is analogous to melting (solid-liquid) phase transition (SL type of PPT).

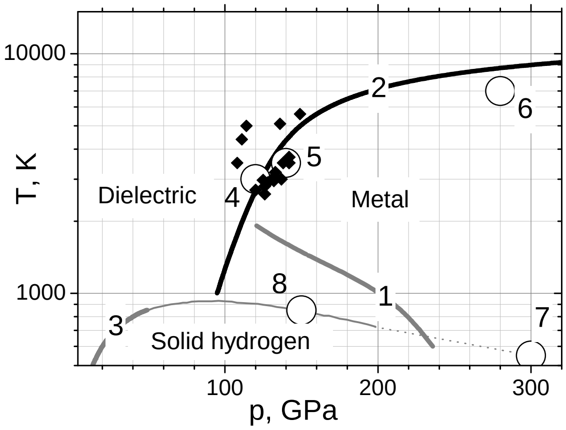

Figure 1. p-T phase diagram of hydrogen. Comments in text.

Curve 1 in the Fig. 1 shows coexisting curve with critical point for PPT of LV type from [5], curve 2 shows coexisting curve for PPT of SL type [3]. Hydrogen melting line 3 from [6] is shown as bold solid (experimentally investigated region), thin solid, and dot line. Points 4 - 8 are experimental data, which can be interpreted, following [3], as states, located in the vicinity of the phase boundary of metallic liquid hydrogen. Diamonds – location of maximum compression states, reached in this study, according to simulation.

The main goal of the presented research was to carry out experiments, capable to prove the existence or absence of the PPT in hydrogen, and show type of PPT, if possible. Experimental results were compared with the results of 1D hydrodynamic simulation of the experimental situation using hydrogen EOS [3].

EXPERIMENTAL PROCEDURE

A series of experiments with different initial pressures of hydrogen was done. Parameters (p, ρ, T) during compression were determined from 1D hydrodynamic simulation with EOS [3] for hydrogen and EOS [7,8] – for other materials.

Initially gaseous hydrogen samples at temperature of about 78 K and pressure varied in the range of 3-30 MPa were used. Experimental assembly, similar to [9], consisted of cylindrical container, cooled by liquid nitrogen and filled by gas of spectroscopic purity. Hydrogen layer with initial thickness of 2 or 4 mm was compressed between stainless steel bottom (thickness of 1.6 mm) and window, composed of lithium fluoride disk, lying on sapphire disk (both are about 5 mm thickness and 20 mm in diameter). Dynamic compression was initiated by the impact of explosively launched stainless steel striker plate (thickness of 4 mm) with container through the 4 – 5 mm layer of aluminum (used to diminish the first shock in H2 and to approach an isentropic compression path). The velocity of striker was near 5.2 km/s. 2-layers structure of window serves to hold initial hydrogen pressure (sapphire layer) as well as to transmit the emission during compression (LiF layer). Our previous experiments with sapphire window [9] and results [10] convinced us of this window choice. The profiles of intensity of optical emission through composite window were measured by fast optical pyrometer in a manner, described in [9].

RESULTS AND DISCUSSION

Experimental brightness temperature profiles for lowest (A), high (B) and intermediate (C) initial pressures of hydrogen, and corresponding profiles from simulation are shown in Fig. 2. They were analyzed on the basis of conductive heat transfer on hydrogen - LiF and hydrogen – steel boundaries taking into account hydrogen phase state and it’s transparency, analogous to [11].

In all the experiments a triangular temperature pike is registered at the end of compression. This pike cannot be explained only by thermodynamics of compression stages (when we should see only a subsequent rise of temperature), and can be explained as being due to formation of thin metal hydrogen film on the cold LiF surface (~1000 K). Metallization of hydrogen leads to abrupt

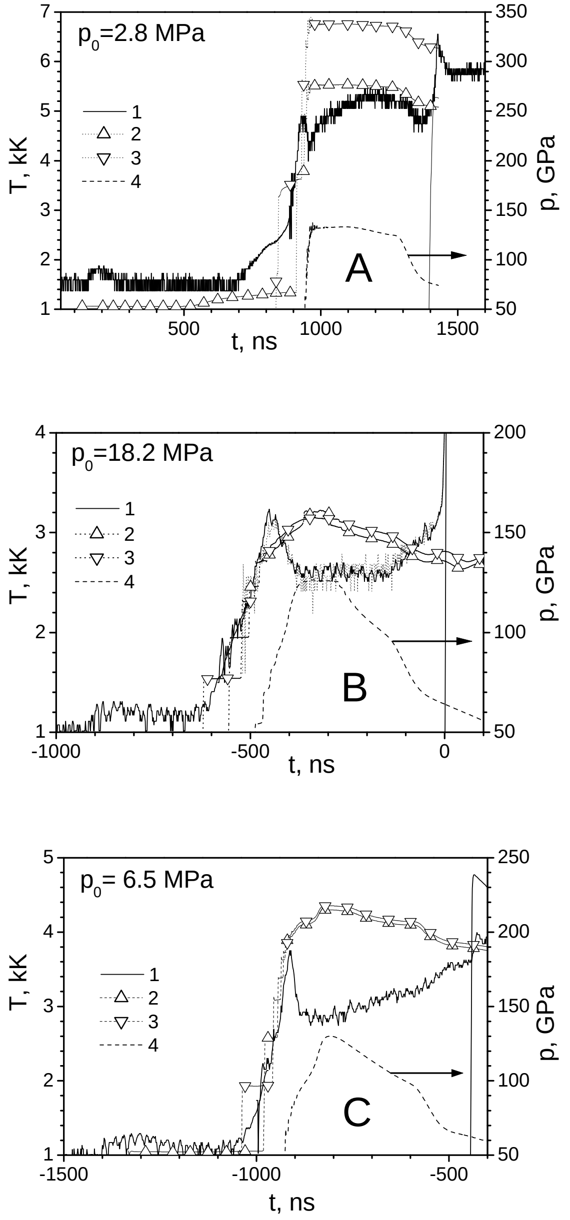

Figure 2. Experimental temperature profiles of hydrogen under compression and results of hydrodynamic simulation with EOS model [3]. 1.Experimental temperature profiles. 2. Model hydrogen temperatures for steel -hydrogen boundary. 3. Model hydrogen temperatures for hydrogen-LiF boundary. 4. Model pressure.

temperature increase according to EOS model [3]. Subsequently, due to the heat transfer this layer is cooled down, resulting in formation of experimentally registered triangular pike at the end of compression. In the experiments with low initial hydrogen pressure and thus high final temperatures (Fig. 3A) this film afterwards disappears. Hydrogen becomes transparent near LiF surface and registered temperature is in agreement with temperature of hydrogen near steel-hydrogen boundary from simulation (Fig. 3A curve 1, 2).

In the experiments with high initial pressure and thus low final temperature (Fig. 3B), a near constant, slightly rising temperature profile is registered after pike in spite of significant pressure and temperature lowering in the bulk of hydrogen. This can be explained by the fact that in this experiments thermal emission from opaque metal hydrogen film on the LiF surface is registered. Time dependent profile for bulk hydrogen temperatures according to model [3] differs significantly from experimental results.

In the intermediate region (experiments with initial hydrogen pressure 5-10 MPa (Fig. 3C), metal hydrogen film with low temperature is also formed after temperature pike. Afterwards, due to the heat transfer from high temperature dielectric hydrogen this film is heated up and subsequently disappears. In this region registered temperatures abruptly change from 3000 K– temperature of metal hydrogen on the LiF surface up to 4000 K – temperature level near the steel-hydrogen boundary.

Curve 3 in the Fig. 3A shows the temperature of the hydrogen front layer on the boundary with LiF window from simulation. This temperature is 1500 K higher then temperature registered in the experiment (curve 1) and is close to the temperature near the hydrogen-steel boundary (curve 2). Thus we can conclude that hydrogen layer near H_2 – LiF boundary remains transparent even at higher temperature, transmitting thermal radiation from steel surface, heated by hot H_2.

In Fig. 3. different symbols are used to represent two groups of experiments, where metal film temperature was measured (temperature is near constant and slightly increasing after pike) and experiments with hydrogen-steel surface temperatures (temperature rises after pike and then diminishes according to the hydrodynamic model). The results are in good agreement with the supposed in [3] PPT for liquid hydrogen.

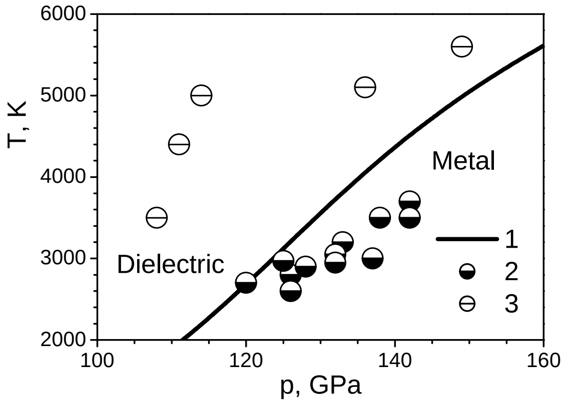

Figure 3. Hydrogen p-T diagram in the region of investigation. 1 - PPT according to [7]; 2 - experimental points with near constant, slightly increasing temperature profiles; 3 - experimental points with registered temperature profiles similar to results of 1D hydrodynamic simulation.

CONCLUSIONS

Final temperature of dynamically compressed hydrogen was investigated for the pressures 100-150 GPa with temperatures being 2500-5500 K. Anomalous temperature pike at the end of compression stage was registered in all the experiments. Registered low temperature profiles shapes differ from that obtained in hydrodynamic simulation. These effects were described by formation of metal hydrogen film on the cold surface of LiF. In high temperature experiments metal film was dissolved. Optical emission of hydrogen with T=6800 K and P=150 GPa was not registered. It appeared to be optically transparent.

Experimentally obtained points in p-T diagram for dielectric and metal states of hydrogen are in a good agreement with the model [3] predictions.

Metal hydrogen film formed on the window material can significantly influence the results of conductivity measurements in the vicinity of phase transition curve.

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