Phase Transition in a Strongly Nonideal Deuterium Plasma Generated by Quasi-Isentropical Compression at Megabar Pressures

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High-explosive driven generators of cylindrical and plane shock waves in D_2 and H_2 were used for the generation of warm and dense strongly nonideal matter with an intense interparticle interaction and Fermi statistics. Highly resolved flash x-ray diagnostics were used to measure the adiabatic plasma compressibility. The thermodynamic measurements demonstrated the 20% increase of density at megabar pressure, just in the density range, where the electrical measurements indicated a sharp—5 orders of magnitude—increase of electrical conductivity due to pressure ionization in strongly coupled plasmas.

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The investigation of Jupiter, Saturn, brown dwarfs, and D-T targets for inertial confinement fusion [1,2] demands the knowledge of thermodynamic information on strongly nonideal (where the ratio of the Coulomb interaction energy to kinetic energy $\Gamma = Ze^2n^{1/3}/E_K \gg 1$), partially degenerated ($n_e \lambda_e^3 \geq 1$; $\lambda_e = \sqrt{2\pi\hbar^2/m_e k_B T}$ ...-electron de Broglie thermal wavelength) hydrogen plasma. In this region the complexity of a physical description increases considerably, when nonideal effects are superimposed with chemical reactions associated with partial pressure and temperature dissociation and ionization [1–3]. Many interesting phenomena, such as metal-insulator transition, Mott effect, and metastability have been predicted for nonideal plasmas. They occur in situations where both quantum and Coulomb effects are important, making the physical analysis extremely difficult. Moreover, new nonstandard kinds of phase transitions have been predicted for strongly coupled Coulomb systems and warm dense hydrogen. It is the so-called "plasma phase transition" (PPT) [1–9] and "dissociation driven" phase transition (DPT) [10–14]. In many of these works [2,5–8,11] the variants of "chemical picture" were applied for calculation of thermodynamics of warm dense partially ionized hydrogen (deuterium).

Recently the *ab initio* direct quantum mechanical simulation approaches [path-integral Monte Carlo method [15] and density functional theory (DFT) molecular dynamic method [12–14]] have been applied successfully to dense warm strongly coupled proton-electron system (hydrogen) and have shown the clear indication of existence of new phase transitions other than the ordinary gas-liquid transition and melting of molecular crystal. That is why the experimental detection of new phase transitions seems to be the real challenge of modern plasma and condensed matter physics. Up to now, no experimental indications of plasma phase transitions in the real multicomponent (ion, electron, atom, and molecule) nonideal plasma were obtained. In this work we present the first direct experimental thermodynamic signature of new phase transition in strongly coupled ($\rho \sim 1$–4 g/cc) nonideal deuterium with intense Coulomb interaction ($\Gamma \sim 100$–200), quasi-isentropically compressed up to pressure ~3 Mbar.

A number of experiments have been made with intense shock compression of liquid, solid, and initially precompressed gaseous hydrogen (deuterium) [16–19]. In early experiments with laser-driven shock waves [16] liquid deuterium was compressed in maximum up to density $\rho \sim 1.1$ g/cc ($\rho/\rho_0 \sim 6$) at pressure range 100–300 GPa. In later experiments with magnetically driven [17] and high-explosive (HE) driven [18,19] shock waves liquid, solid, and initially precompressed gaseous deuterium was compressed up to significantly lower density $\rho \sim 0.7$–0.8 g/cc ($\rho/\rho_0 \sim 4$) at pressure 100 GPa. All these experiments gave no indication of any new phase transitions. Present experiment gives such indication at the same pressure as the shock experiments ($p \sim 100$ GPa) but at significantly higher density ($\rho \sim 2$ g/cc).

To generate strongly nonideal plasma the dynamic method was applied (see [1] and references therein). In this experimental approach the intense shock waves were used for compression and nonreversible heating of plasma up to several megabars pressure range and up to temperatures ≤ 3–8×10^3 K. The inertial confinement of high-pressure plasma allows us to generate strongly coupled states during 10^{-8}–10^{-6} sec.

In our experiments we have used the quasi-isentropic compression of plasma [20–23] by the series of reverberating shock waves, allowing one to obtain compression $\rho/\rho_0 \sim 50$ and $\Gamma > 200$.

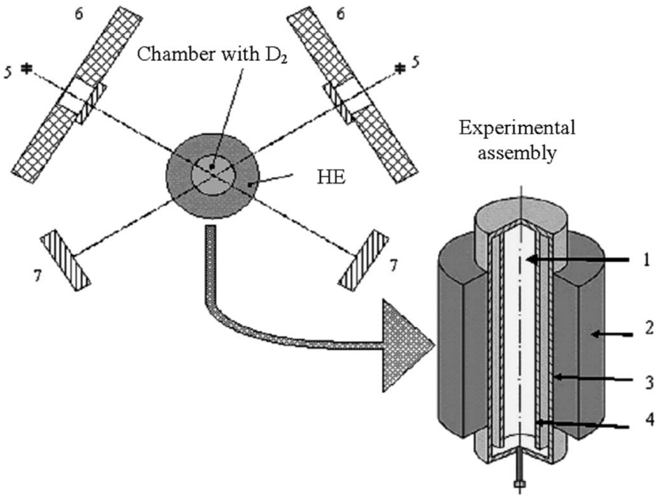

FIG. 1. Experimental assembly for quasiadiabatic compression of deuterium plasma.

The scheme of the experiment is presented in Fig. 1. To dynamically compress the high-pressure deuterium in cylinder 1 the cylindrical high-explosive charge 16, 24, and 32 kg were used. The HE charge was ignited simultaneously in 640 points by the special detonation optic device. Compression of deuterium plasma in sample 1 was performed by the steel liners 3 and 4, accelerated by the high enthalpy detonation products of HE charge 2 and by the reverberating shock waves in the volume 1.

The pulsed x-ray radiography of the explosive experimental device was performed by two independent hard x-ray sources 5 protected by the heavy metal protection system 6. X-ray photo of the devices was registered by the analog-to-digital converter (ADC) "Agfa" with high ($\sim 10^4$) dynamic range, high sensitivity, and fine spatial resolution. Screens 7 were used to increase the quantum efficiency. The technique of the special computer procedure of x-ray picture statistical analysis allows us to detect the position of interface between liner and deuterium plasma with standard deviation equals to -0.08% and $+0.37\%$. The static (a) and dynamic (b) x-ray experimental device pictures with densitygram (c) and (d) are shown in Fig. 2. The comparison of the 2D computer simulation of deuterium plasma cylindrical compression with x-ray measurements of inner liner position (Fig. 3) demonstrates a good agreement between simulations and observation. Equations of state from [24] and elastic-plastic model from [25] were used in the simulations.

The sets of 2D computer simulations of HE compression of the experimental assembly, using different 2D computer codes, different equations of state, and elastic-plastic models, have shown that the pressure of multiple shock compressed deuterium can be obtained with the accuracy $\leq 3\%$. The obtained experimental data are presented in

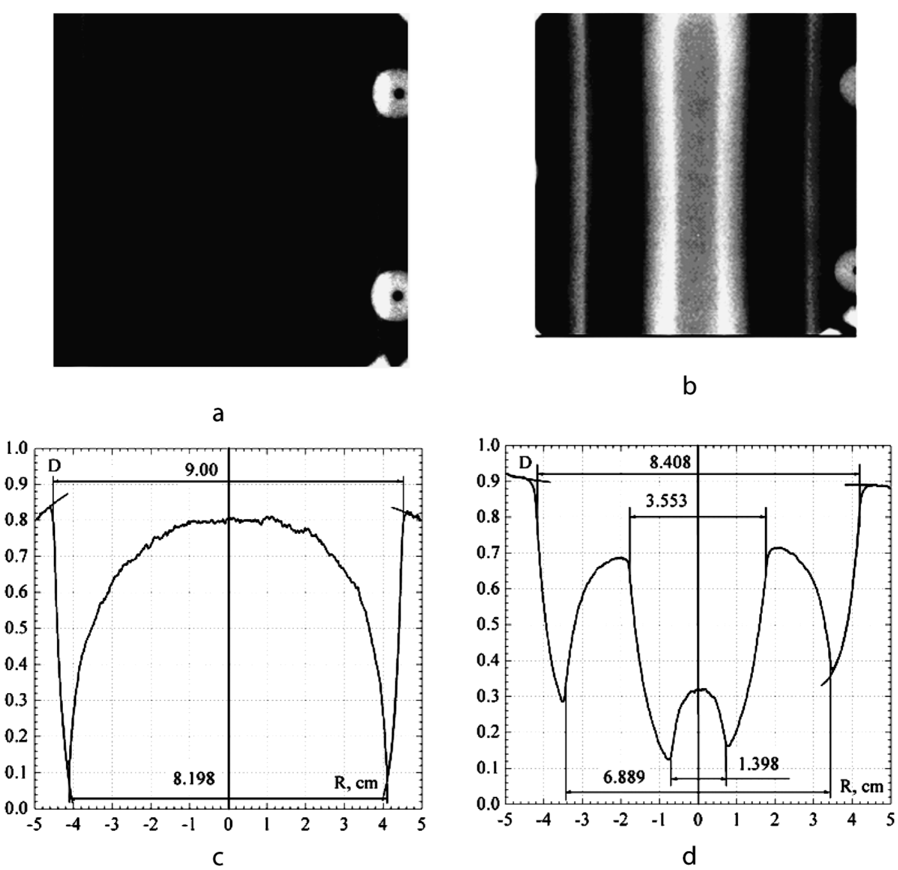

FIG. 2. X-ray picture of experimental sample before (a) and during dynamic compression, after 43.04 μs from ignition (b); (c), (d)—optical density (D) of x-ray film (a) and (b) as a function of distance (R) from axis (0) of cylinder.

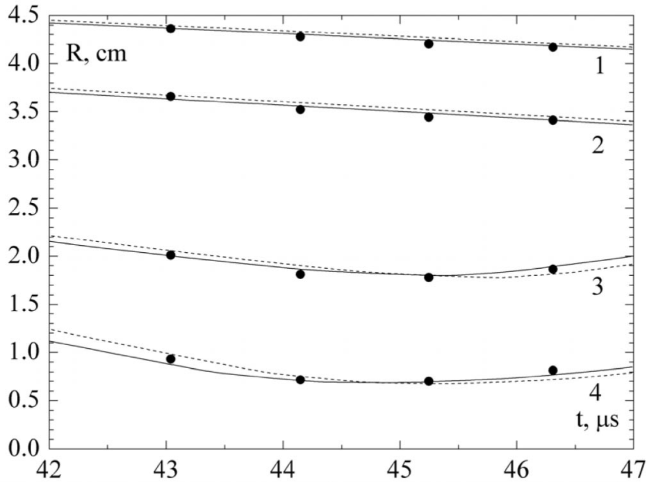

FIG. 3. Trajectories of the experiments for quasi-isentropic deuterium compression up to $P = 125$ GPa. Points—x-ray measurements; 1, 2—external and 3, 4—internal liners positions; solid and dashed lines—hydrodynamic simulation with different elastic-plastic models [25].

the Table I, where R_0 and ρ_0 are the initial radius of inner liner 4 and initial deuterium densities.

X-ray measurements were performed during the period of existence of states with maximal achievable density. According to hydrodynamic simulation this period is about 2 μs, much more than duration of x-ray pulse (300 ns). The density of quasi-isentropically compressed plasma $\rho_{\rm exp}$ was obtained with the accuracy 3%–5% from inner liner position $R_{\rm min}$, detected by the flash x-ray, and pressure—from hydrodynamic simulation, based on experimental information on inner liner position.

In Table I and Fig. 4 the experimental data obtained are compared with some theoretical models of nonideal plasmas [21,24,26] and experiments [27], which were performed more then 30 years ago. It is seen that a reasonable agreement between experimental data [27] and our present measurements exists, except one experimental point from [27] designated as K_1.

The experimental data obtained (Fig. 4) demonstrate good agreements with experiments [27] and theoretical models [21,24,26] in the pressure range up to 130 GPa. At higher pressures 125–130 GPa, our experiments demonstrate the density "step" in the nonideal deuterium adiabatic compressibility—the registered density of deuterium at pressure 140 GPa (point 3, Table I) is 25% higher than density at $P = 127$ GPa (point 2). It should be noted that the density jump appearance corresponds to a strong increase of plasma nonideality parameter (up to $\Gamma_D \sim$ 100–200) and electron degeneracy effects in dense deuterium plasma.

The additional information on density anomalies in nonideal plasma was obtained from reverberating shock compression experiments in plane geometry [20,21]. In these experiments the registration of time-resolved electrical and optical signals from shocks in plasma at high pressure has shown much longer than predicted shock wave propagation time intervals and thus the higher plasma density at high Γ regimes.

To understand the experimental results, the combination of the "confined atom" model [28] with "chemical picture" approach [26] has been developed (SAHA-D model). This model reproduces two fragments on isentrope with initial pressure and density corresponding to the lowest experimental point (lowest star at Fig. 4). It is remarkable that the high density branch of calculated isentrope proved to be in good agreement with two high-pressure experimental points (two right stars at Fig. 4). The plasma parameters calculated with SAHA-D code, which correspond to experimental points, are presented in Fig. 4 and Table I.

The direct path-integral Monte-Carlo method (see [15] and references therein) and molecular dynamic simulation superimposed with the density functional theory (DFT) ([12,13,21] and references therein) clearly demonstrates the thermodynamic anomalies and phase transition just in the same density range where our experiments were performed (Fig. 4). Ability of these two approaches (PIMC and DFT-MD) to reproduce presently measured parameters of deuterium isentrope is an open question.

We think we have obtained the first direct experimental signature of the existence of new phase transition in strongly nonideal hydrogen (deuterium) with intense Coulomb interaction other than the gas-liquid transition and melting of molecular crystal. It is remarkable that strongly coupled nonideal plasma thermodynamic anomalies, registered here by the x-ray techniques, correspond to

TABLE I. Experimental results on deuterium plasma compression in comparison with equation of state models calculations.

No	R_0, mm	$R_{\rm min}$, Mm	ρ_0, g/cc	$\rho_{\rm exp}$, g/cc	P, GPa	$\rho_{\rm calc}$ g/cc [24]	$T_{\rm calc}$, K, [24]	$\rho_{\rm calc}$ SAHA-D [26]	$T_{\rm calc}$, SAHA-D [26]	$\Gamma_{\rm calc}$, SAHA-D [26]	$n_e \lambda_e^3$ SAHA-D [26]
1	54	10.2 ± 0.13	0.0404	1.09 ± 0.03	77	1.156	3100	1.09	2180	0.02	7.8×10^{-8}
2	41	6.99 ± 0.12	0.0396	1.36 ± 0.05	127	1.420	3850	1.29	2500	135	8.4
3	54	7.82 ± 0.16	0.0386	1.78 ± 0.07	150	1.509	4100	1.36	2600	150	9.6
4	54	6.87 ± 0.17	0.0396	2.35 ± 0.12	255	1.896	5200	$\cdots$	$\cdots$	$\cdots$	$\cdots$
5	40	5.07 ± 0.14	0.0396	2.46 ± 0.14	302	2.065	$\cdots$	2.52	2410	594	146

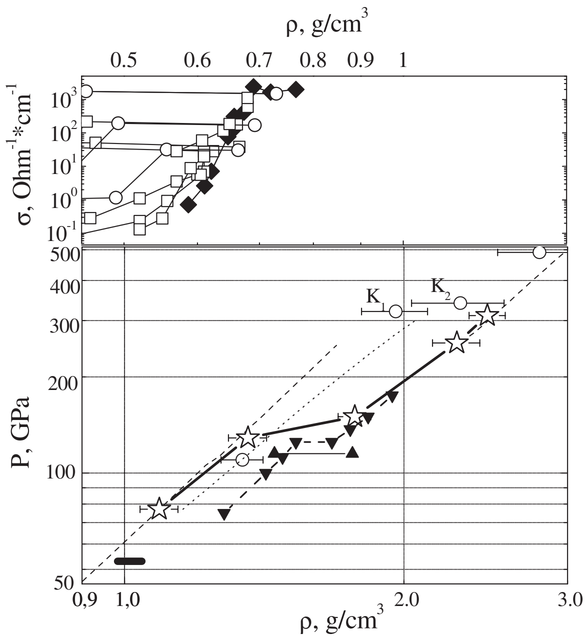

FIG. 4. Isentropic compression of deuterium. Comparison of experimental data and theoretical predictions. Open circles—[27]; line with stars—present work; dashed line isentrope calculated by SAHA-D model [21,26]; dotted line—isentrope calculated by model [10,24]; hypothetical phase transitions: density gap at "plasma phase transition" (PPT) at $T = 2000–10\,000$ K [6]—up triangles; "dissociation driven" phase transition (DPT) at $T = 1500$ K [12]—down triangles with dashed line; anomaly at $T = 3000$ K and $\rho = 1$ g/cm^3 associated with DPT [13]—thick line; Insertion above. Electrical conductivity of compressed hydrogen: solid rhombus—data from experiments with liquid hydrogen [20,21]; open circles and open squares—data from experiments with liquid and gaseous hydrogen accordingly [22,23].

a sharp—4 orders of magnitude—jump (see insertion in Fig. 4) of dc electrical conductivity of adiabatically compressed plasmas. We plan to extend both the experimental and theoretical techniques to search and study phase transitions and electrical conductivity anomalies in warm dense matter of other elements and at other parameters.

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