Experimental measurements of the compressibility, temperature, and light absorption in dense shock-compressed gaseous deuterium

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Introduction

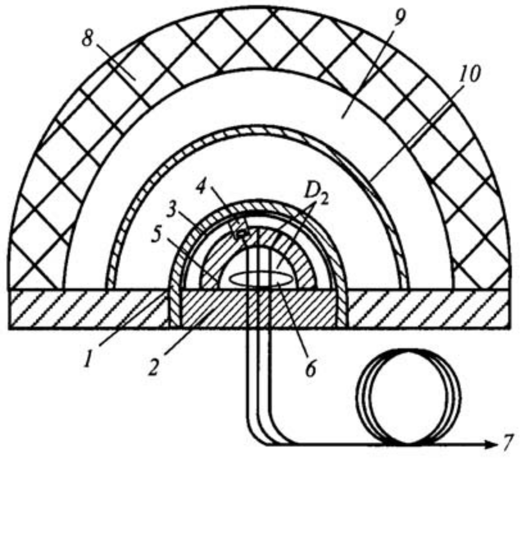
At present, advances in laser fusion and progress in the understanding of the structure and evolution of astrophysical objects have quickened interest in the study of thermodynamic and electrophysical properties of hydrogen - the simplest and most abundant element in nature - in the megabar pressure range. To achieve shock-compression megabar pressures, various methods of shock-wave excitation are used: intense laser radiation [1], high-power pulse currents [2], and spherical explosion devices [3-4]. Although the laser data demonstrate anomalously high compressibility of deuterium plasma, this was not confirmed by the electrodynamic and explosion experiments.

In this work, we studied properties of gaseous deuterium with a high initial density, close to the density of liquid deuterium. The temperature and light absorption coefficients were measured simultaneously with the compressibility. It allowed additional information to be gained on the parameters of the state and optical properties of the shock-compressed deuterium plasma.

The use of gaseous deuterium was dictated by the possibility of obtaining its initial parameters with a high certainty, because they are fully determined by the initial gas pressure and temperature.

Experiment

For the experiments with gaseous deuterium under a high initial pressure, a hemispherical capsule was devised. It is schematically shown in fig. 1.



Capsule frame (1) and base (2) were made from a high-strength steel possessing high stability of its characteristics in a hydrogen atmosphere. To enhance the shock-compression pressure in deuterium, hemispherical 1.5-mm-thick aluminum (AD-1) screen (3) was placed under steel frame (1).

Fig. 1. Hemispherical experimental device: (1) frame; (2) base; (3) screen (aluminum AD-1); (4) aluminum (AD-1) sample; (5) housing; (6) optical sensors; (7) measuring line; (8) explosive; (9) air gap; (10) impactor (steel 3).

At a fixed distance (determined by the height of three aluminum samples (4) from the aluminum screen, hemispherical brass housing (5) was mounted, inside which 13 optical sensors (6) for measuring shock velocity in deuterium were symmetrically arranged. Similar sensors were also placed beneath the samples (4) to measure shock velocity in them and use this velocity for determining the shock-compression parameters in the aluminum screen. The sensors were fabricated from 200-μm o.d. silica fibers with several-micron-thick aluminum jackets along the whole length to eliminate spurious illumination.

The fibers (6) were glued in base (2) that provided tightness of the construction, and in brass hemisphere (5); their polished top ends were mounted flush with the outer surface of the hemisphere. The bottom (in the scheme) ends terminated in the optical connector (not shown in the figure) for joining the optical sensors to external fiber lines (7), through which the shock-front radiation was transmitted to detectors. The central fiber, with a diameter of 600 μm, served also for measuring the shock velocity in gas and the shock-front temperature.

The capsule was filled with gaseous deuterium from a metal-hydride source of high-pressure vanadium-based hydrogen isotopes. To achieve a pressure of 250 MPa using the vanadium-deuteride source, it suffices to heat it to a temperature of ~ 450 K.

Two experiments, with the initial gas parameters P_0 = 203 MPa (2000 atm) and T_0 = 273 K in the first experiment and P_0 ~ 157 MPa (1550 atm) and T_0 = 278 K in the second, were performed using shock-wave generators of the same type. Under these conditions, the initial gas densities calculated according to [5] were ρ_0 = 0.153 g/cm^3 and ρ_0 =0.1335 g/cm^3, respectively.

The shock-front glow was detected in the visible region by optical transducers based on photodiodes with a signal rise time no worse than 2 ns and photo-multipliers with an anode-pulse rise time of 1.2 ns. The shock-wave passage time was measured from the instant of glow appearance to the instant of glow decay due to the damage of the fiber end by the shock wave. The typical oscillograms of the shock-front glow in gaseous deuterium are shown in Fig. 2 (a, b). The brightness temperature of shock-compressed deuterium was determined from the glow amplitude *h* (Fig. 2(b)) in the saturation region.

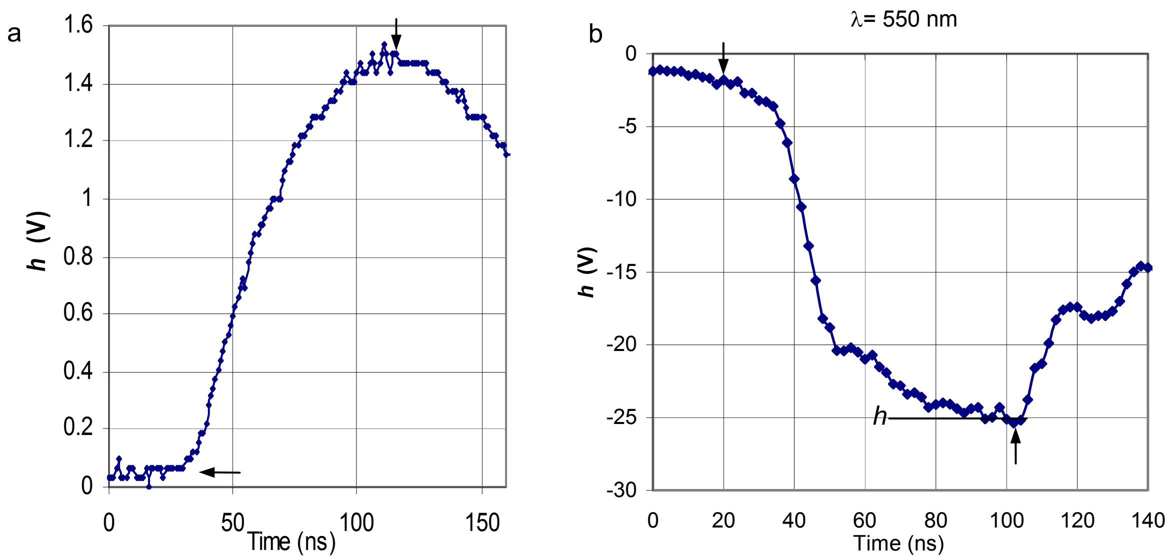


Fig. 2. Oscillograms for shock wave front radiation in gaseous deuterium. Detectors: (a) photodiode sensors and (b) photomultipliers. Arrows show the points at which the time of shock-wave motion was measured

The measurements of the average shock velocities in dense gaseous deuterium gave D_{exp} = (29.14 ± 0.56) km/s at $P_0 \approx$ 203 MPa (2000 atm) and D_{exp} = (29.29 ± 0.36) km/s at P_0 = 157 MPa (1550 atm) [4]. The experimental values of average velocity of shock wave in deuterium corresponded to the measurement radius R_{meas} = 1.53 cm. The average velocity of shock wave i aluminum samples D_{Al} = 21.28 km/s was measured in two experiments at the same radius.

Comparing to [4], in this work, the obtained data on deuterium compressibility are reprocessed. For processing of the results, we used the new equation of state of aluminum [6] and the isentrope of aluminum unloading (ρ_o = 2.71 g/cm^3) from the initial state on shock adiabat with parameters D_{Al} = 21.28 km/s, U = 12.606 km/s, and P = 726.95 GPa. Besides, the measured velocities of shock waves were not corrected. The problem on decay of random break at measurement radius was solved. In this case, at the indicated velocities, the use of the law of conservation of mass bring about the following shock-compression parameters for gaseous deuterium: *D*=29.14 km/s, *U*=22.82 km/s, *P*=102 GPa, ρ=(0.705 $\pm$ 0.06) g/cm^3 in the first experiment and *D*=29.29 km/s, *U*=23.2 km/s, *P*=91 GPa, ρ=(0.64 $\pm$ 0.04) g/cm^3 - in the second experiment.

The temperature was measured using a high-speed optical pyrometer for visible range of spectrum. The radiation of the shock front in deuterium was recorded via the fiber line at wavelengths of 450, 498, 550, and 600 nm. To separate the corresponding spectral intervals, a set of interference light filters with a transmission bandwidth at half $\Delta\lambda \approx$10 nm was used. Prior to the experiments, the optical line for measuring temperature was calibrated against a reference light source. The thermal radiation flux from a heated body with the radiating capacity *Э* is given by the Planck's formula:

$$N(\lambda) = Э\, C_1 \lambda^{-5} \left[\exp\left(C_2 / \lambda T\right) - 1\right]^{-1} \tag{1}$$

Here, *Э* is the radiating capacity of the body, λ is the wavelength, T is the actual temperature, and the constants are C_1 = 1.19·10^{-16} (W m^{-2})/sr and C_2 = 0.0144 mK. The temperature of the shock-compressed gaseous deuterium was determined from the four measured spectral temperatures by the nonlinear least-squares method for two parameters T and *Э,* followed by iterations to obtain exact estimates for the quantities of interest. At the spectral temperatures experimentally measured for the shock-compression pressure *P = 93* GPa in the range 450-600 nm, the best fit to the Planck's function is achieved, in the grey-body approximation, at the temperature *T*=24100±2200 K and a radiating capacity *Э* of 0.485±0.075. In the experiment, saturation of dependence of shock wave front radiation, which would correspond to optical thickness of shock-compressed gaseous deuterium close to unit, was not reached at pressure P = 83 GPa. The average value T = 22900 $\pm$ 2000 K of the brightness temperature was presented for this experiment.

The rise of the shock-front glow after the wave enters gaseous deuterium can be due to the increase in the thickness of the shock-compressed layer and to its transparency. Then, with the known kinematical parameters *D* and *U,* one can use the experimental oscillogram to determine the light absorption coefficient α in the shock-compressed deuterium with thickness *l*:

$$\alpha = -\left[1/(D-U)t\right]\ln(1 - N/N_0), \tag{2}$$

where No – radiation amplitude in the saturation zone, N – radiation amplitude at time t. For (4), light reflection was neglected, and transmittance was determined using the Bouguer-Lambert-Beer formula:

$$\tau = \exp\left(-\alpha l\right), \tag{3}$$

The average value of this coefficient obtained for the compressed gaseous deuterium after processing the experimental oscillograms was found to be $\alpha \approx$ 69 cm $^{-1}$ in the wavelength range 450-600 nm for a shock compression of 93 GPa.

Comparison with the results of theoretical calculations

The experimental results are presented in Fig. 3 in the pressure-density coordinates and, in the temperature-pressure coordinates, in Fig. 4, together with the available experimental data and the results of theoretical calculations in two variants.

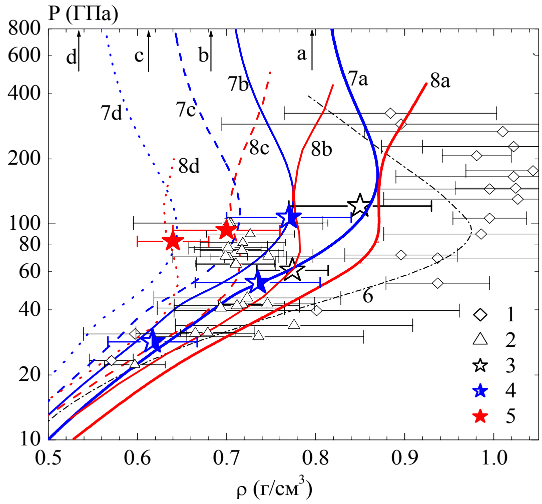


Fig. 3. Deuterium Hugoniot adiabates. Experiment: 1 – [1], 2 – [2], 3, 4 – [3], 5 – this work. Calculations: 6 – [1], 7 - SAHA-IV [4], 8 - CCM [4], solid thick line for ρ_0 = 0.199 g/cm^3, solid thin line for ρ_0 = 0.171 g/cm^3 , dash-and-dot line for ρ_0 = 0.153 g/cm^3 , and the dotted line for ρ_0 = 0.1335 g/cm^3. Arrows indicate the "limiting" compressions (ρ / ρ_0 = 4) for each of the four Hugoniot curves.

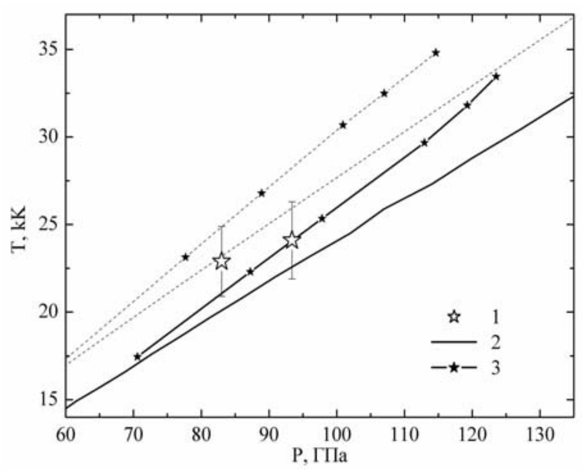


Fig. 4. Pressure dependence of the temperature of shock-compressed gaseous deuterium: (1) experiment; calculation: (2) SAHA-IV model [this work], (3) CCM model [7]; solid lines are for ρ_0 = 0.153 g/cm^3 and dashes are for ρ_0 = 0.1335 g/cm^3.

In the first variant, the calculations were carried out using the equation of state constructed for hydrogen within the relatively simple compressible covolume model (CCM) [7]. A mixture of five sorts of particles was considered: molecules, atoms, positive molecular ions, protons, and electrons. The thermal equation of state for the particle of the *i*-th sort has the form $V_i(P,T) = V_{C,i}(P)+RT/P$, where V is the molar volume and R is the molar gas constant. The covolumes $V_{C,i}$ were assumed to depend only on pressure and be additive. For molecules, the covolume was constructed using the experimental data on the static compression of solid hydrogen up to a pressure of 2.5 GPa [8] and quasi-adiabatic compression of gaseous hydrogen in the pressure range 40-800 GPa [9]. For molecular ions and protons, they were taken to be formally equal to the covolumes of, respectively, molecules and atoms (for electrons, $V_{C,e}(P)\equiv 0$). The caloric equation of state was obtained from the thermal equation using the second law of thermodynamics. The contribution from the vibrational and rotational degrees of freedom of molecules and molecular ions in the partition functions was taken into account in the "rigid rotator-harmonic oscillator" (RRHO) approximation, the contribution from the excited electronic states was disregarded. For the states achieved in our experiments, the CCM calculations give $n_D/n_{D2} \sim 3$ and relatively low degree of “temperature ionization”. The CCM-calculated Hugoniot curves show two density maxima: the lower corresponds to the molecular dissociation, and the upper corresponds to the particle ionization. The position of the second maximum can be affected by many factors. In particular, use of RRHO approximation for molecules and molecular ions especially causes significant growth of density and corresponding pressure at high temperatures.

In the second variant, the calculations were performed using the universal SAHA-IV code [10]. In this model, hydrogen (deuterium) was calculated as a strongly non-ideal mixture of ions, electrons, atoms, molecules (and negative and molecular ions). When calculating the equilibrium plasma composition and its thermodynamic properties, the partial degeneracy of the electronic component and the interactions between all sorts of particles were taken into account. To describe the coulombic nonideality, an improved modification of the pseudopotential approach was used. Apart from the contribution from the Coulomb interaction between charged particles, the strong repulsion of heavy particles at close distances was taken into account. This was accomplished using the approximate equation of state for "soft spheres", modified to a mixture of particles with different diameters. In our calculations, the parameters of this interaction were chosen according to the non-empirical “atom-atom approximation”.

Calculations in the modified plasma chemical model show that the states arising behind the shock-wave front in this work and in the experiments with maximal pressure from [3-4] correspond to a dense strongly nonideal ($\Gamma >> 1$), partially ionized ($n_e/n_H \sim 1$), partially degenerate ($n_e\lambda_e^3 \sim 3$), and practically isothermic ($T \approx$ 22-24 kK) deuterium plasma.

An analysis of the data [3-4] presented in the table shows that the physical conditions realized in the shock experiments at RFNC-VNIIEF are distinguished by a combination of strong coulombic non-ideality ($\Gamma >> 1$), electron-component degeneracy ($n_e\lambda_e^3 \sim 1$), and strong influence of the short-range repulsion that manifests itself in the high values of the so-called packing parameter $\pi n_e D_e^3/6 \sim 1$ (здесь D_i – is the self-size of a heavy particle of the *i*-th sort (atom, molecule, etc.)).

Complete comparison of the experimental data with calculation results is presented in Fig. 3. One can see that the experimental data obtained in this work agree well with the data calculated on the basis of the CCM and SAHA-IV models. It is worthy of note that these theoretical models *agree simultaneously* both with the results of our experiments on the shock compression of a preliminarily compressed gaseous deuterium and with the results of the shock compression of liquid and solid deuterium [3]. This suggests that the results of all shock-wave experiments conducted at RFNC-VNIIEF are mutually consistent. Nevertheless, the fact that, although the Hugoniot adiabates in both theoretic cal models tend to the ideal-gas asymptotic compression limit $\rho_{Hug}/\rho_0 \rightarrow 4$ at the high-pressure and high-temperature limits, the character of this tendency is different for both models. This renders the necessity of obtaining new experimental data in the pressure range ~ 2-10 Mbar topical and necessitates the use of *ab initio* calculations for determining the thermodynamic functions of dense hydrogen (deuterium) plasma.

The estimates made for the absorption coefficient within the framework of the classical approach using the calculated parameters and the Kramers-Unsold formula give $\alpha \approx 4{\cdot}10^4$cm^{-1}, which is three orders of magnitude larger than the measured values. Such a discrepancy is evidence that the ionization and dissociation processes at the shock front likely bypass the rise of the plasma-bunch radiation.

Conclusions

1. Results of the performed work show that experimental values of density and temperature are in good agreement with data of the calculations performed by the CCM and SAHA-IV models.

2. The developed design of the experimental device can be used also for study of properties of gaseous deuterium cooled down to temperature of, at least, liquid nitrogen. In this case, the initial density of gas will be higher than density of liquid deuterium.

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