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EXPERIMENTAL DETERMINATION OF THE COMPRESSIBILITY OF HYDROGEN AT DENSITIES 0.5 - 2 g/cm<sup>3</sup>. METALLIZATION OF HYDROGEN

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The equation of state of hydrogen is of interest because hydrogen is the main component of certain planets and of most stars. Furthermore, it can go over into a metallic superconducting state at high pressures.

A theoretical analysis of the equation of state of hydrogen has been the subject of many papers [1 - 7]. All predict the existence in solid hydrogen (at  $T = 0^\circ\text{K}$ ) of a transition from the molecular to the atomic modification, at densities on the order of 0.5 - 1 g/cm<sup>3</sup>. In this density region, an energywise more favored state is one in which the molecules are broken and the hydrogen becomes a monovalent metal. The phase-transition pressure fluctuates in the theoretical estimates in the range 0.25 - 18 Mbar.

There are few known studies of the compressibility of hydrogen. Stewart [8] measured the isotherm of solid molecular hydrogen at  $T = 4^\circ\text{K}$  up to pressures  $P = 20$  kbar. Mills and Grilly [9] determined experimentally its melting curve up to  $T = 60^\circ\text{K}$  and  $P = 3.5$  kbar. Van Thiel and Alder [10], in experiments on shock compression of liquid hydrogen ( $\rho_0 = 0.071$  g/cm<sup>3</sup>), reached a density 0.19 g/cm<sup>3</sup> at 39.5 kbar.

We have measured by a gamma-graphic method the density of hydrogen in the course of its isentropic compression by a cylindrical shell in the pressure range from 0.4 to 8 Mbar. We used for this purpose a cylindrical explosive charge, the products of which accelerated a shell that compressed gaseous hydrogen to high densities and pressures. Devices of this type are described in [11 - 14]. To register the diameter of the cavity

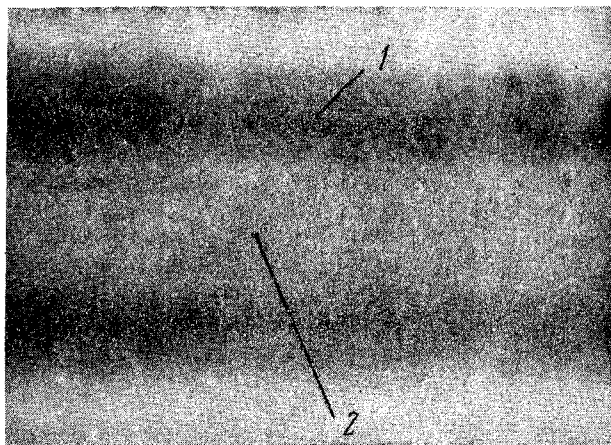


Fig. 1. Experimental gamma-diagram: 1 - metallic shell, 2 - cavity with compressed hydrogen.

Experimental data

№	$\rho$ , g/cm <sup>3</sup>	Meas. error $\Delta\rho/\rho$ , %	$\delta = \rho/\rho_k^{1)}$	P, Mbar
1	0.45	12	5,1	0.37
2	0.98	17	11,0	2.63
3	1,15	18	12,9	3,24
4	1,40	21	15,7	4,40
5	1,95	40	21,9	8,00

<sup>1)</sup>  $\rho_k = 0.089$  g/cm<sup>3</sup> is the density of H<sub>2</sub> at T = 0 and P = 1 bar.

with compressed hydrogen, we used a high-power transmission gamma-graphic installation with a small exposure time, of the type described in [15]. A characteristic gamma-diagram of the shell cavity at one of the instants of compression of the hydrogen is shown in Fig. 1. By measuring the diameter of the cavity with the compressed gas during the process of convergence of the shell to the axis, one can determine the density of the hydrogen. By varying the initial pressure of the gas and the parameters of the discharge, it is possible to determine the compressibility of the hydrogen in a sufficiently wide range of densities. The experimental results are listed in the table.

The table shows also the values of the pressures obtained from gasdynamic computer calculations, using the equation of state of hydrogen obtained by the method described above.

On the basis of the experimental data obtained in this and earlier known experiments, we determined the equations of state of the solid and liquid phases of molecular hydrogen. The form and method of finding the parameters of the equation of state were assumed to be similar to those in [16 - 18].

The results of the calculation of the isentrope corresponding to the experiment, of the absolute-zero isentrope ( $\rho_x(\rho)$ ) obtained with the contribution

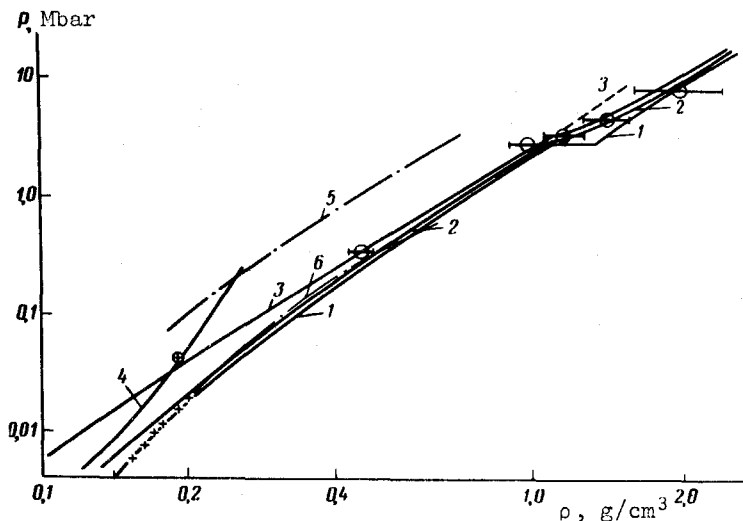


Fig. 2. Experimental and calculated plots for hydrogen: barred circles - our experimental results, crosses - Stewart's experimental isotherm at  $t = 4^\circ\text{K}$  [8]; crossed circles - shock-compression experiment [10].  $P(\rho)$  - zero isotherm (1), melting curve (2), isentrope (3), shock adiabat (4), all calculated in the present paper; (5) -  $P_x(\rho)$  according to Abrikosov [4]), (6)  $P_x(\rho)$  according to Trubitsyn [6].

of zero-point oscillations subtracted, of the melting curve, and of the shock adiabat from the initial state with  $\rho_0 = 0.071 \text{ g/cm}^3$  are shown in Fig. 2. It is seen from this figure that the calculated isentropes and the shock adiabat agree quite satisfactorily with the experimental ones. A similar agreement is obtained for the calculated melting point over the entire experimentally investigated region.

We call attention to the fact that, according to experiment, hydrogen at  $P \approx 0.4 \text{ Mbar}$  has a density  $0.45 \text{ g/cm}^3$ , which is approximately 1.8 times larger than the value calculated by Abrikosov [3] for the molecular phase, and is sufficiently close to Trubitsyn's calculation [6]. This is natural, since the attraction terms in the pressure were not taken into account in [3].

The difference between the calculated isentrope of the molecular phase at  $P > 2.8 \text{ Mbar}$  (dashed curve in Fig. 2) with experiment points to the existence of an anomaly in the properties of hydrogen in this pressure region. This anomaly can be identified with the transition of the hydrogen into the metallic state.

All the available experimental data can be satisfactorily described by assuming that the transition into the metallic modification of hydrogen in the solid state occurs at a pressure  $P = 2.8 \text{ Mbar}$ , with a density jump from  $1.08$  to  $1.3 \text{ g/cm}^3$ .

The elastic-interaction curve  $P_x(\rho)$  for atomic hydrogen coincides here with the zero isotherms calculated by different authors at  $P \geq 30 \text{ Mbar}$  [3, 5], and differs somewhat from them at lower pressures.

Calculation of the temperature on the isentrope and on the melting curve at pressures  $\sim 3 \text{ Mbar}$  shows that the anomaly takes place in the liquid phase. Assuming that the transition occurs with a zero entropy jump, the phase-equilibrium curve crosses the melting curve at  $T \approx 700^\circ\text{K}$ . Since the calculated temperature on the isentrope at these pressures is  $\sim 7000^\circ\text{K}$ , it is apparently impossible to identify the kink on the experimental isentrope with the solidification of the hydrogen upon compression.

We note that calculations of the isentrope and of the melting curve at  $P > 2.8 \text{ Mbar}$  were performed under the assumption that there exist two liquid phases of hydrogen, between which a second-order phase transition exists<sup>1)</sup>.

The calculated isentropes and the melting curve, calculated from the obtained equations of state, are shown in Fig. 2.

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<sup>1)</sup>An analogous anomaly takes place in liquid NaCl following shock compression [19].

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#### DEGENERACY OF EXCITON GAS FOLLOWING POWERFUL OPTICAL EXCITATION IN CdS CRYSTAL

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The question of producing non-equilibrium exciton gas of high and controllable density is still a pressing one. We have attempted to determine by direct measurement the occupation numbers of the excitons produced in CdS crystals by powerful single-photon excitation. At the maximum pumps attained in our experiment ( $P \sim 3 \times 10^6$  W/cm<sup>2</sup>), these numbers turned out to be  $n \geq 1$ .

To determine the exciton occupation numbers, we used a method consisting of measuring the gain in the region of the induced exciton-phonon luminescence [1]. The exciton occupation numbers were then estimated directly from the values of this coefficient.

It was shown earlier [1] that an important factor limiting the accumulation of the exciton in the band during optical pumping in CdS crystals is their induced radiative decay. This decay can be limited by decreasing the dimensions of the excited region of the crystal. In our experiment, one of the dimensions of this region was determined by the absorption coefficient for the exciting light ( $L_1 \sim 1/\alpha \sim 10^{-4} - 10^{-5}$  cm), while the other two dimensions, which determined the area of surface excitation, could be controlled under the conditions of the experiment ( $L_2 \leq 10^{-2}$  cm,  $L_3 \geq 10^{-3}$  cm). Using a tunable pulsed dye laser [2] delivering a maximum power density  $\sim 10^6$  W/cm<sup>2</sup> at a generation line width 2 - 3 cm<sup>-1</sup> and a monopulse duration 10 nsec, we excited "monochromatic" excitons in a wide spectrum of their motion energy  $\epsilon(\vec{k})$  (up to  $\epsilon(\vec{k}) \sim 3\hbar\omega_{LO}$ , where  $\hbar\omega_{LO}$  is the energy of the optical phonon). We used high-grade CdS crystals in which the donor and acceptor densities did not exceed  $5 \times 10^{14}$  cm<sup>-3</sup>, and the edge-dislocation density was less than  $10^2$  cm<sup>-2</sup>. The investigations were performed at  $T = 77^\circ\text{K}$ , when radiative decay processes of only free excitons were manifest in the spectrum [3]. Figure 1 shows the exciton spontaneous emission spectrum at a low excitation power level,  $\sim 10^{-2}$  W/cm<sup>2</sup> (A corresponds to direct radiative annihilation of the excitons and A - LO corresponds to the "red" boundary of the exciton-phonon transitions with emission of LO phonons). The induced exciton-phonon luminescence corresponding to a maximum linear dimension of the excited region of the crystal on the order of  $10^{-2}$  cm occurs at single-photon excitation power densities close to  $10^4$  W/cm<sup>2</sup> (curve 2 of Fig. 1). Up to  $10^6$  W/cm<sup>2</sup>, processes of radiative decay of free excitons predominate in the induced-luminescence spectrum (curves 3 - 7). There was no generation of light, since the induced luminescence had a single-pass character<sup>1)</sup>.

<sup>1)</sup>Lasing on exciton-phonon transitions in CdS was first observed in [4,5].