Features of low-temperature plasticity of solid hydrogen

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A substantial dependence of the rate of the steady-state creep of H_2 at helium temperatures on the ortho-para composition of the samples has been observed. When the concentration of the ortho modification is decreased to 0.2%, the rate of deformation of the hydrogen increases by more than 50 times.

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Interest in the investigation of the plasticity properties of crystals at low temperatures has increased in recent years. As shown in[1–3], the behavior of a crystal under load can be qualitatively entirely different in this case, owing to the influence exerted by quantum effects on the plastic-deformation processes. In view of the small mass of the molecules and the relatively weak van der Waals interaction between them in the lattice, the influence of quantum effects should be particularly large in the case of solid helium and hydrogen. Investigations of the plasticity of hydrogen in the interval 1.4–$4.2\,°K$[4] have led, however, to the conclusion that its deformation down to $1.4\,°K$ is determined by the thermally-activated motion of the dislocations. The low values of the activation energy of the process ($U < 20$ cal/mole) have made it possible to suggest that the plastic deformation of H_2 at helium temperatures is connected with the overcoming of the Peierls barriers by the dislocations. Attention is called, however, to the fact that the rate $\dot{\epsilon}$ of the hydrogen deformation in the orientationally-disordered phase ($T > 1.6\,°K$) turns out to depend on the ortho-para composition of the samples. A decrease of the concentration of o-H_2 from 75 to 2% has led, in the entire investigated temperature interval, to more than a twofold increase of $\dot{\epsilon}$. Taking into account the sensitivity of a number of kinetic properties of H_2 at low temperatures to the presence of small amounts of the ortho impurity,[5–7] we have investigated the plasticity of hydrogen in the interval 2–$4\,°K$ with samples in which the o-H_2 content was reduced to 0.2%.

We investigated the rate of plastic deformation of p-H_2 under uniaxial tension of the samples by a constantly applied load. In a wide temperature and stress interval, we also investigated the plasticity of normal hydrogen (75% o-H_2). All the measurements were made on perfectly transparent polycrystalline samples ($l = 30$ mm, $d = 6$ mm) obtained from the liquid phase and subsequently removed from the glass-cell walls by pumping vapor over the sample. The results obtained for solid n-H_2 at a stress $\sigma = 5.5$ g/mm^2 ($\sigma/G = 5 \times 10^{-4}$, G is the shear modulus) are shown in Fig. 1. Analysis shows that at $T > 9\,°K$ the temperature behavior of the rate of the steady-state creep of n-H_2 is described by the usual thermal activation dependence with $U = 200\,°K$, which coincides with the self-diffusion activation energy.[8] It has been established that in this temperature region $\dot{\epsilon}$ has a power-law dependence on σ with an exponent $n = 3$–3.5. The obtained data indicate that the plasticity of hydrogen at $T > 9\,°K$ is deter-
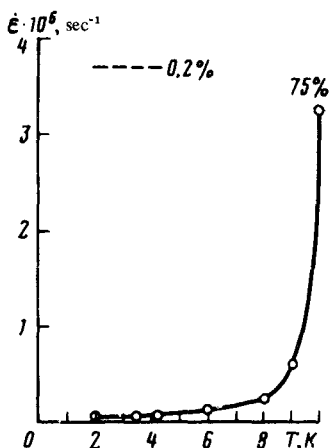
FIG. 1. Dependence of the rate of the steady-state creep of H_2 on the temperature. Curve—samples of normal hydrogen, dashed—samples of para-hydrogen with 0.2% ortho-modification.

mined by one of the diffusion mechanisms whereby the dislocations are released from the blockades.

At helium temperatures, the rate of deformation of solid n-H_2 depends relatively little on the temperature, the dependence being linear at low values of the deforming stress ($\sigma < 15$ g/mm^2). The values obtained here for the rates agree well with the data of[4]. Measurements performed at helium temperatures on samples from which the ortho-admixtures have been eliminated have revealed anomalously high values of $\dot{\epsilon}$. The value of $\dot{\epsilon}$ turned out in this case to be quite sensitive both to the quality of the samples and to the presence of extraneous impurities in them (the initial purity of the H_2 was not worse than 99.9999%). The limiting value of $\dot{\epsilon}$ for p-H_2 at $\sigma = 5.5$ g/mm^2, established from the investigation of 35 samples, is 3.68×10^{-6} sec^{-1}, which is more than 50 times the value obtained for n-H_2. Despite the high quality of the crystals, this value was reached only in several experiments. For a number of samples, an increase of $\dot{\epsilon}$ was noted only in the range 200—300%. Our mass-spectrometric analysis revealed unexpectedly a rather clear-cut correlation between $\dot{\epsilon}$ and the content of deuterium in the samples. It was established that the deuterium accumulates in the liquid p-H_2 during the course of its purifica-
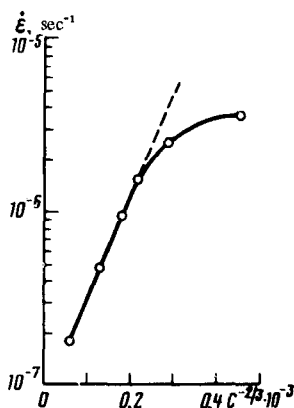

FIG. 2. Dependence of the rate of steady-state creep of para-hydrogen on the concentration of deuterium.

tion of the ortho-admixture. The minimum value obtained for p-H_2 is $\dot{\epsilon} = 1.75 \times 10^{-7}$ sec^{-1}, which corresponds to a deuterium concentration $C_{D_2} = 0.22\%$. The experiments have shown that at a given C_{D_2} the rate of deformation in the interval 2—4 °K is practically independent of temperature. It can be concluded that the principal role in the processes of the detachment of the dislocations from the D_2 molecules is played not by thermal fluctuations but by quantum fluctuations. In this case[9]

$$\dot{\epsilon} \sim \exp \left[(U - \gamma\sigma) / T_{eff} \right], \tag{1}$$

where U is the height of the barrier preventing the dislocation motion, γ is the activation volume, and T_{eff} is the effective temperature. At low temperatures, T_{eff} can be set equal with sufficient accuracy to a constant. The effect exerted on the rate of deformation of the D_2 impurity is obviously determined by the $\gamma(C_{D_2})$ dependence. At low deuterium concentration γ, and consequently $\ln\dot{\epsilon}$, should be proportional to $C_{D_2}^{-2/3}$. As follows from Fig. 2, this relation is satisfied down to $C_{D_2} = 0.03\%$. Assuming that the plastic deformation of the hydrogen is connected with slipping in the basal planes, and using the corresponding exact expression for γ, we obtain from the slope of the $\ln\dot{\epsilon} \sim C_{D_2}^{-2/3}$ line that $T_{eff} = 12.7$ °K. This agrees well with the value $T_{eff} = 13$ °K obtained from the formula[9]

$$T_{eff} \approx T_m \, \Delta_0^2 / \Delta_m^2 , \tag{2}$$

where T_m is the melting temperature and Δ_0^2 and Δ_m^2 are the mean-squared displacements of the H_2 molecules at 0 °K and at T_m. At $C_{D_2} < 0.03\%$ the experimentally observed velocity is apparently determined by the presence in the sample of a residual o-H_2 impurity (0.2%). The mechanism that limits the mean free path of the dislocations in the crystal can in this case be their stopping their ortho-molecule clusters. It is known that the electrostatic quadrupole-quadrupole interaction of the ortho-molecules, being the nearest neighbors in the lattice, exceed by 4 °K the para-molecule interaction energy.[10] At low o-H_2 concentrations and accordingly at large dislocation velocities, the clusters are more readily overcome because the dislocations lose a part of their kinetic energy. At o-H_2 concentrations, the principal role is obviously assumed by activation processes. Extrapolation to 0 °K of the temperature dependences of the activation energy in n-H_2 and p-H_2 with 2% of o-H_2, obtained in[4] for several values of σ, leads to respective values 4 ± 0.2 and 4.5 ± 0.5 °K, which coincides in fact with the energy that characterizes the interaction of the ortho-molecules in the cluster.

Thus, our experiments did not reveal any influence of the Peierls relief on the rate of plastic deformation p-H_2. In parahydrogen that is perfectly free of impurities, the rate of deformation can apparently reach exceedingly high values. The question of whether this velocity is reached as a result of above-barrier motion of the dislocations or of coherent tunneling of the dislocations through the Peierls barriers calls, however, for further study. In this respect, the investigation of ϵ as a function of σ is of interest. Anomalous $\dot{\epsilon}(\sigma)$ dependences should be observed in the region of low stresses. If the stress applied to the crystal leads to a collapse of the dislocation levels in the valleys of the Peierls relief.

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