

Surface diffusion of adsorbed molecular hydrogen at low temperatures

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(Submitted 17 July 1985)

Pis'ma Zh. Eksp. Teor. Fiz. **42**, No. 6, 253–255 (25 September 1985)

The temperature dependence of the surface diffusion coefficient of molecular hydrogen adsorbed on the tungsten (110) face has been studied at low temperatures. The results imply a tunnel mechanism for the hydrogen diffusion.

The fortunate combination of a small mass of the hydrogen molecule with a relatively weak binding with the close-packed (110) face of a tungsten single crystal suggests that the surface mobility of adsorbed hydrogen at low temperatures, which has been observed previously, is due to quantum diffusion.¹ In this letter we report an attempt to determine the surface diffusion coefficient D of hydrogen as a function of the sample temperature and the defectiveness of the surface of the tungsten crystal.

Data on the mobility have been obtained in a study of the kinetics of structural phase transitions in adsorbed films. The study was carried out by the method of the static skin effect,^{2,3} which is based on the effect of the adsorption on the characteristics of the surface scattering of conduction electrons in metals.^{4,5}

The vacuum chamber, the preparation of the single-crystal tungsten wafers [the (110) face; thickness ~ 0.1 mm], and the technique used to clean them are all described in Ref. 5. The measurements are carried out in high vacuum ($p \sim 10^{-10}$ torr) at a temperature close to the temperature of liquid helium. A signal proportional to the resistance of the sample is fed to the input of a differential amplifier and to an S8-13 oscilloscope. The wafer temperature is set by the current flowing through the sample and is measured by a differential W-Re(5%)/W-Re(20%) thermocouple. A static magnetic field ($H \sim 10$ kOe) is imposed in the plane of the wafer, perpendicular to the measuring current.

Figure 1 shows traces of the magnetoresistance $\Delta\rho(t)$ of a tungsten wafer as a function of the exposure time in a hydrogen atmosphere. The parameter of the curves is the hydrogen pressure; the sample temperature is 8.3 K. The traces are nearly bell-shaped, tending toward saturation in the vicinity of a monolayer coverage ($\theta = 1$). It has been shown previously¹ that the inflection point on the $\Delta\rho(t)$ curve at the lower part of the figure corresponds to a structural phase transition $p(2 \times 2) \rightarrow p(2 \times 1)$. We see that a gradual increase in the hydrogen deposition rate (the central and upper curves) erases the structural feature associated with the formation of the $p(2 \times 1)$ phase and increases the plateau height, which characterizes the defectiveness of the latter structure, $p(1 \times 1)$. For estimates we assume $D = \langle \Delta x \rangle^2 / \tau$, $\Delta x \sim 3 \times 10^{-8}$ cm, and $\tau = 0.1 t_0$, where t_0 is the minimum time required for the formation of the $p(2 \times 1)$ structure.^{1,6} Figure 2 shows the results of estimates found at various temperatures. We

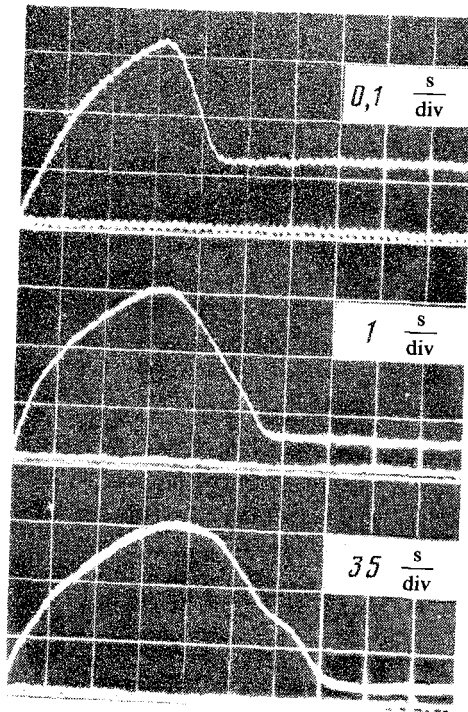


FIG. 1. Magnetoresistance of a tungsten wafer as a function of the exposure time in a hydrogen atmosphere. The lower trace corresponds to a pressure $p \sim 10^{-6}$, the middle trace to 10^{-5} , and the upper trace to 10^{-4} torr.

see that the anomalous temperature dependence of the diffusion coefficient, $D(t)$, indicates a mechanism of resonant tunneling of the molecules.^{7,8}

According to Kagan and Maksimov,⁸ the conditions for resonant tunneling may be disrupted near the static defects which are unavoidably present on a surface, with the result that a complete localization of the particles occurs at high defect concentra-

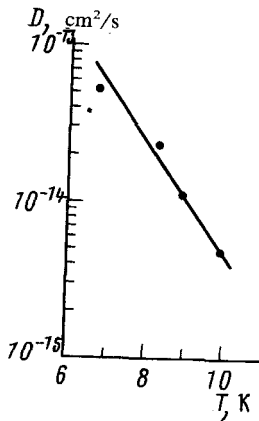


FIG. 2. Temperature dependence of the surface diffusion coefficient of hydrogen.

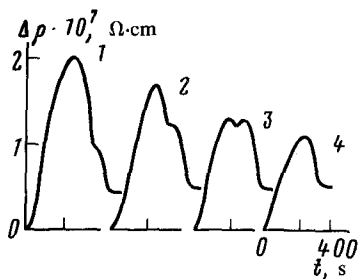


FIG. 3. The $\Delta\rho(t)$ dependence for various surface defect concentrations. 1—The cleanest possible crystal surface; 2, 3, 4—after the preliminary deposition of 0.03, 0.12, and 0.30 of an oxygen monolayer.

tions. Data supporting this mechanism are shown in Fig. 3. The surface is made defective through the deposition of small portions of oxygen. The first $\Delta\rho(t)$ curve corresponds to the cleanest surface possible by our technique. The impurity concentration increases for the other $\Delta\rho(t)$ curves: 0.03θ for curve 2, 0.12θ for curve 3, and 0.30θ for curve 4, where $\theta = 1$ corresponds to an oxygen monolayer. The hydrogen pressure during these measurements was $p \sim 10^{-6}$ torr, and the crystal temperature was $T = 8.3$ K. We see that curves 1 and 2 are quite different, despite the fact that the average distance between oxygen atoms is $\sim 15 \text{ \AA}$. For an average distance of $\sim 7 \text{ \AA}$ (curve 3), we find a nearly complete localization. This localization prevents the development of the $p(2 \times 1)$ phase, which is "specular" in terms of electron reflection. A further increase in the impurity concentration leads to the formation of a structureless hydrogen film in the region of submonolayer coverages, but the $p(1 \times 1)$ structure is preserved in the limit $\theta \rightarrow 1$.

A further detailed study of the temperature and isotopic dependence of D will apparently yield a definitive answer to the question of the nature of the diffusion of molecular hydrogen.

We wish to thank R. Sh. Nasyrov for furnishing the pure tungsten samples.

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Translated by Dave Parsons