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Hydrogen current generator based on Pd/InP Schottky diode

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A new current generator based on Pd/InP Schottky diode with an area of 1 mm² operating in hydrogen medium in complete darkness has been created. It is established that in a short-circuit regime a “hydrogen current” of the Schottky diode increases by about three orders of magnitude with increasing the temperature from 90 to 300 K. It is shown that the increase of the “hydrogen current” with increase the temperature is related to the decrease of the potential barrier height of the diode in the temperature range 90–300 K. The mechanism of current generation in Pd/InP Schottky diodes in 100 % hydrogen, consisting in appearance of an additional number of electrons in Pd-layer as a result of palladium hydride formation, is proposed.

Keywords: hydrogen, palladium, Pd/InP, ionization, palladium hydride.

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Introduction

Electrical properties of Schottky diodes Pd/InP in the gaseous environment with hydrogen are studied to create hydrogen detectors [1–5]. At the same time, changes in current-voltage and capacity characteristics of such diodes are studied in hydrogen compared to air environment, and all measurements are usually performed when applying an external voltage. In hydrogen, the work function of palladium changes [6, 7] and, consequently, the height of potential barrier in Pd/InP diodes, at the same time, the electrical characteristics of diodes also change, and the change of characteristics can be used to judge the hydrogen concentration in gaseous environment.

Previously, current conduction mechanisms [1, 2] in Pd/InP structures in hydrogen environment with and without LED illumination of the structure at room temperature without the application of external voltage were investigated. In study [3] current generation was detected in Schottky diodes Pd/InP when placing them in hydrogen environment without applying external bias. This study is a continuation of the above-

mentioned papers and dedicated to the study of current generation and EMF in Pd/*n*-InP structures in the temperature range of 90–300 K in the hydrogen atmosphere.

Experiment

The manufacturing process of the studied Pd/InP structures is described in detail in [1–3]. A palladium layer with an area of 1×1 mm² and thickness of 25 nm was created by thermal vacuum deposition of 99.96 % purity on InP single crystal wafers with the electron concentration Pd of 10¹⁶ cm⁻³. Gold-based ohmic contacts were created for palladium and InP. Electrical properties of Pd/InP structures were studied in the temperature range of 90–300 K in vacuum and hydrogen environment. The study of structures was conducted under the conditions of short-circuited electrical circuit and in the open-circuit mode. To assess the temperature dependence of the height of the potential barrier in the studied Schottky diodes, current-voltage characteristics were measured in complete darkness in vacuum and in the 100 % hydrogen environment.

When studying electrical characteristics of Pd/InP structures, KEITLEY-6517B (Keithley Instruments, Inc.) electrometer was used with data output to a computer.

Experiment results

Let us consider the current behavior in the hydrogen atmosphere in the temperature range of 90–300 K (fig. 1). “Hydrogen current”, i.e. the current that appears only in hydrogen environment in the short-circuit mode in complete darkness, increases by about 3 orders of magnitude with temperature increase from 90 to 300 K.

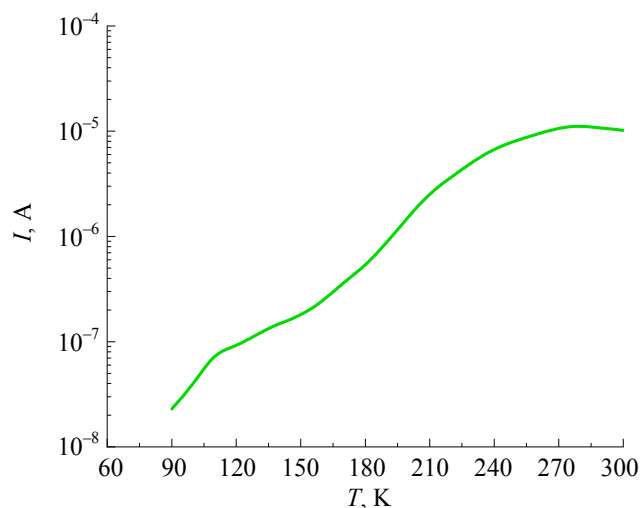


Fig. 1. Temperature dependence of current generation in the hydrogen atmosphere

At $T \sim 270$ K, the current reaches the maximum value of $10 \mu\text{A}$ at a structure area of 1 mm^2 . Based on the dependence of EMF (E) on the temperature in the hydrogen environment (fig. 2), fluctuation in the considered temperature range can be distinguished, however, there is a tendency for EMF to increase in absolute value when temperature decreases. EMF reaches the maximum value $|E| = 170 \mu\text{V}$ at a temperature of 90 K.

Fig. 3 shows the dependence of the potential barrier height of Schottky diode Pd/InP on the temperature in 100 % hydrogen. Height of the potential barrier was determined according to the cutoff potential value of the current-voltage characteristics extrapolated to $I = 0$.

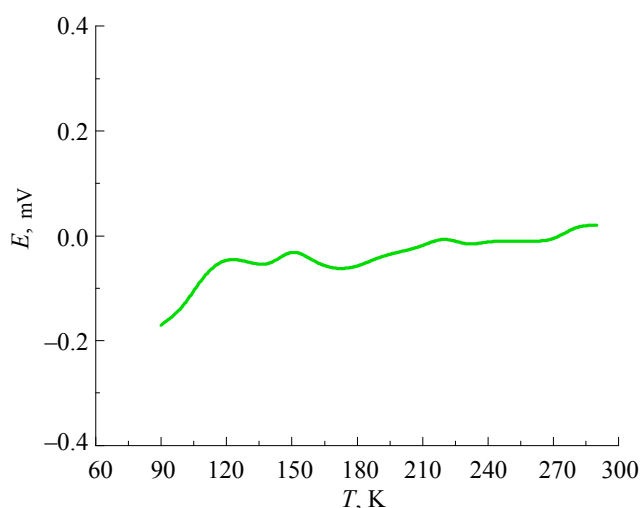


Fig. 2. Temperature dependence of EMF generation in the hydrogen atmosphere

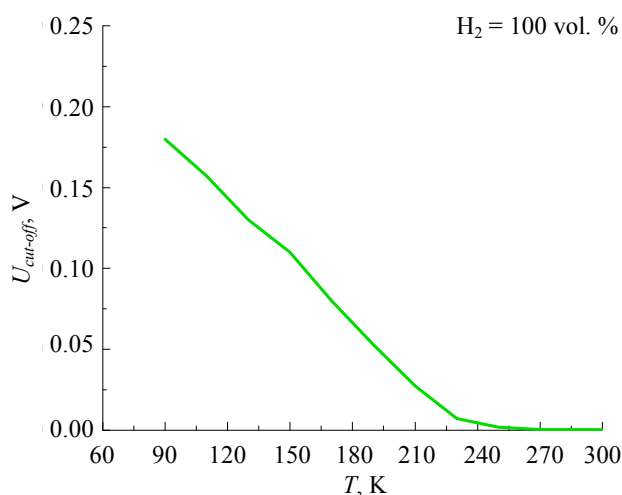


Fig. 3. Temperature dependence of the potential barrier height

The presented chart shows that the barrier height of Schottky diode in the hydrogen medium decreases monotonously from 0.18 to 0 eV with increase of the temperature from 90 to 270 K.

Results analysis

The appearance of “hydrogen current” in Schottky diodes in the hydrogen environment can be related, according to the authors, to the formation of an additional electrons in the Pd-layer. Formation of the additional number of electrons, in turn, can be the result of two factors. Firstly, ionization of hydrogen atoms is possible upon contact of hydrogen with Pd-layer. As was shown in study [8], when hydrogen atoms approach Pd atoms at a distance less than Bohr radius, energy of hydrogen atoms ionization can be reduced to 0 eV. Secondly, upon contact of hydrogen with Pd-layer, palladium hydride PdH_x is formed [9, 10]. The process of PdH_x formation is exothermic and occurs with the release of thermal energy [11]. Combined effect of these two factors possibly leads to the appearance of the additional electrons in the palladium layer.

After the formation of the additional electrons, they drift from the palladium layer to InP. Potential barrier of Schottky diode significantly restricts the flow of electrons formed at the result of ionization. In the hydrogen environment

(100 vol. %), the barrier height of Schottky diodes Pd/InP decreases linearly from 0.18 to 0 eV when temperature increases from 90 K to 270 K. At the barrier height close to 0 eV, conditions arise for free electrons flow along the electrical circuit. The current generated as the result of this effect is maximum at a temperature of 275–280 K. With a further increase of temperature to 300 K, a gradual decrease in the generated “hydrogen current” begins. The resistance of the potential barrier determined by the slope of current-voltage characteristics at a voltage value of 10 mV decreases from 10^5 to 1–2 Ohm at temperature increase from 90 to 300 K.

Fig. 4 shows band diagrams explaining the obtained experimental result. The height of Schottky diode Pd/InP barrier in vacuum is determined by difference between the work function of Pd and InP and is 0.32 eV at

$T = 90$ K. When palladium interacts with hydrogen in complete darkness at room temperature, ionization of hydrogen atoms occurs with the formation of protons and electrons, and palladium work function W_{Pd} decreases by 0.3–0.7 eV [2, 6].

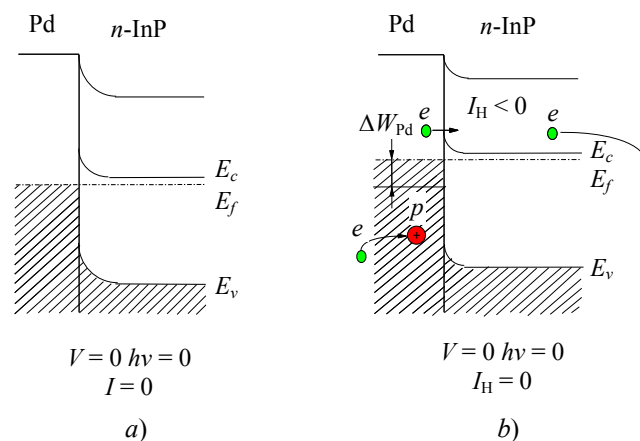


Fig. 4. Band diagrams and currents in the Pd/InP structure, external voltage on the sample is equal to zero, electrical circuit is short-circuited, $T = 90$ K: a – in the vacuum; b – in the hydrogen; I_H – “hydrogen current”; ΔW_{Pd} – change of Pd work function in hydrogen; e – electrons; p – protons

A decrease in W_{Pd} results in decrease in Schottky barrier height. As a result of separation of charge carriers, short circuit current appears in the structure the value of which depends on the barrier height. As the temperature decreases, the barrier height increases and electrons begin to accumulate near the Pd/InP interfacial area. This causes the short-circuit current to decrease and the open-circuit EMF to increase. Duration of current flow in the structure at $T = 300$ K is maintained for a day and then it linearly decreases for 7 days to 10 % of the original value. When hydrogen is repeatedly filled into the cell, the current increases to the same value as at first hydrogen filling (10 μA). The value of generated current (10 μA) is proportional to the structure area (1 mm^2). It is worth noting that with temperature increase from 90 to 300 K, the barrier height in the hydrogen environment will reduce and approach to 0 eV due to the reduction of energy band gap E_g in InP.

Conclusion

The current generation and EMF were studied in vacuum and hydrogen environment in

the temperature range of 90–300 K in complete darkness. In the hydrogen environment, Schottky diodes generate current in the temperature range of 90–300 K. The value of generated current increases by about 3 orders of magnitude with temperature increase from 90 to 300 K. EMF value increases by absolute value from 0 to 170 μV with temperature decrease from 300 to

90 K. Current generation may be due to the formation of the additional electrons due to the formation of palladium hydride during hydrogen interaction with Pd layer.

The authors declare that they have no conflict of interests.

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