

PRESSURE DEPENDENCE OF THE T_C OF SUPERCONDUCTING METAL-HYDRIDES, MEASURED IN A DIAMOND ANVIL CELL

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For the preparation of high concentration metal-hydrides a very large chemical potential of molecular hydrogen is needed. This can be achieved by means of a high pressure diamond anvil cell. We developed a diamond anvil cell technique for filling the sample space at cryogenic temperatures with molecular hydrogen, loading metal samples with hydrogen at high pressures and in-situ measurements of the resistance and superconducting transition of the sample.

1. INTRODUCTION

Use of high hydrogen pressures makes it possible to synthesize high concentration hydrides of metals that are poor hydrogen absorbers. Especially interesting are the high concentration hydrides of Palladium-Noble Metal alloys. These hydrides can reach superconducting T_C 's of ~ 17 K (1) at high enough hydrogen concentrations.

After the discovery of these relatively high T_C 's, theory predicted even higher transition temperatures than were measured (2). Experiments by Antonov et al. (3) however suggested that the high T_C 's are an artefact of the implantation technique used in ref. (1) to load the samples. Recent theoretical work (4,5) did not resolve the controversy.

We developed a diamond anvil cell technique for the synthesis of high concentration hydrides under thermodynamic equilibrium with the surrounding molecular hydrogen and in-situ measurements of T_C as a function of pressure. In this paper we focus our attention on the experiments. A detailed account of the experimental results together with calculations of the electron-phonon coupling parameter will be published elsewhere (6).

2. EXPERIMENTAL

The experiments are performed in a Diamond Anvil Cell (DAC) made of a hardened Beryllium Copper alloy (BerylCo25). With this DAC we can apply a maximum force of 3.10^4 N on the diamond anvils. This makes it possible to use relatively large diamonds and still reach high pressures ($P < 100$ GPa). The DAC is similar to that

of Silvera and Wijngaarden (7). The major differences are in the force applying mechanism and the inclusion of a continuous flow heat exchanger for operation with liquid helium or liquid nitrogen. The force is applied by a lever arm which is displaced by an excentric cam which in turn is moved by a worm gear. The heat exchanger is mounted in the body of the cell and makes rapid temperature sweeps possible with low helium consumption.

Since hydrogen is not only part of the sample but also the pressure transmitting medium we need it at a sufficient starting density. For this we cool down the DAC and fill the sample space with liquid hydrogen. The setup for condensing hydrogen is shown in figure 1. It shows a cross-section of the area around the diamonds where the high pressure is generated. The diamonds are fixed in brass rings using an epoxy resin. A teflon ring surrounds the brass rings. In this way a small chamber is formed around the diamond tips. Gaseous hydrogen is introduced in the chamber through the capillary. When the temperature of the chamber (i.e. the DAC) is just above the triple point temperature of H_2 (~ 14 K) and the leakrate of the chamber is small enough for the pressure to rise above the triple point pressure of hydrogen (~ 0.070 bar), the hydrogen condenses and fills the chamber. The sample and the condensation process can be viewed through the windows of an optical cryostat and the diamonds using a videocamera mounted on a microscope. After condensation of the hydrogen a force is applied to the diamonds to seal the sample space and to pressurize the sample.

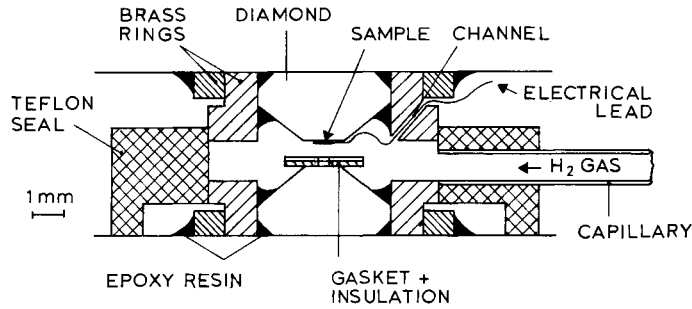


FIGURE 1

Schematic drawing of the setup around the diamonds for condensing hydrogen in the sample space. After condensation the diamonds are pushed together.

To let the hydride formation take place we have to warm up the DAC. Above a certain temperature, depending on the sample material, the diffusion of hydrogen in the sample becomes fast enough to load the sample with hydrogen. The absorption of the hydrogen can be monitored by measuring the resistance of the sample. The resistance of a Pd sample during the loading with deuterium is shown in figure 2. The resistance is measured while continuously heating the DAC. The technique used to measure the resistance of a sample in the DAC is described elsewhere (8).

3. RESULTS

Using the described technique we made high concentration hydrides and deuterides of Pd and a Pd-Ag alloy. At a pressure of ~ 30 kbar we synthesized stoichiometric PdH and PdD for the first time in a diamond anvil cell. The resistance of a Pd-sample during loading with hydrogen or deuterium (see figure 2) shows a large peak. The strong increase around 125 K is due to additional electron scattering, resulting from the random distribution of hydrogen (deuterium) on the octahedral interstitial sites of the Pd host lattice.

At zero pressure we find for PdH $T_c = 8.8$ K and $d \ln T_c / dP = -0.0073$ kbar $^{-1}$ and for PdD we find $T_c = 11.0$ K and $d \ln T_c / dP = -0.0049$ kbar $^{-1}$. For a Pd₉₃Ag₇-hydride (deuteride) we get a concentration of $\sim 85\%$ at loading pressures of ~ 80 kbar. The T_c 's and dT_c/dP 's are similar to those of stoichiometric PdH and PdD. An analysis of the T_c and its pressure dependence using the Allen and Dynes formula for T_c (9,2),

$$T_c = \frac{\langle \omega \rangle_{\log}}{1.20} \exp \left(- \frac{1.04 (1+\lambda)}{\lambda - \mu * (1+0.62\lambda)} \right)$$

shows that the pressure dependence is mainly determined by the volume dependence of the phonon frequencies and not by the volume dependence of the electronic contribution to the electron-phonon coupling parameter λ . The agreement between theory and experiment is excellent (6).

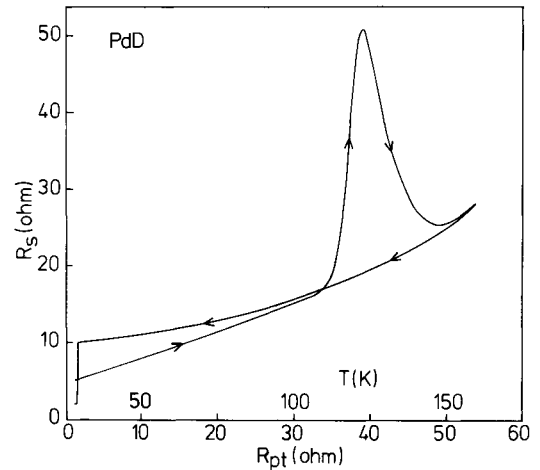


FIGURE 2

Resistance R_s of a Pd sample during loading with deuterium.

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