

Magnetic properties of scandium hydrides

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The first investigation of the magnetic susceptibility of scandium hydrides is reported. The behavior of the magnetic properties in the single-phase region is explained using a proton model.

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Scandium is the first member of the group of transition elements, and its d shell has one electron. It is therefore a convenient object for the investigation of the role of d electrons in the formation of the magnetic properties. In one of the latest calculations of the electronic structure of scandium^[1] it was shown that the Fermi boundary (FB) in this element is at the maximum of the state-density curve. An investigation of alloys with elements having an electron concentration different from that of scandium can

confirm this conclusion. To this end it is necessary to have alloys from the solid-solution region. Particularly interesting are compounds with hydrogen, since hydrogen in compounds with transition metals behaves, in a limited concentration region, like a metal and forms solid solutions with metallic properties.¹⁾ When hydrogen interacts with scandium,³⁾ the first to be produced (up to 30 at.%) is a hydride with hcp structure. This is followed by a region of a mixed state of phases $\alpha + \beta$, which exist up to 67 at.% H. Next to be formed is scandium dihydride (ScH_2), which has an fcc structure. The resistivity of Sc-H in the hcp-structure region is approximately double that of pure scandium.⁴⁾ Scandium dihydride exhibits no superconductivity down to 2 K.⁵⁾ The results of an investigation of the processes of diffusion of hydrogen in scandium by the NMR method⁶⁾ are treated in the proton model, in which hydrogen gives up a fraction of its charge to the conduction band of the metal. Investigations⁷⁾ of the Knight shift and of the spin-lattice relaxation rate (SLRR) in scandium hydride have also confirmed that hydrogen enters the interstices in the form of protons. The SLRR data point to the filling of the d -band of scandium and to an abrupt decrease of the density of states in the hydride.

In the present study we investigated the magnetic properties of compounds of hydrogen with scandium. The original scandium was obtained by triple distillation in vacuum. The hydration was at 600 °C directly in a hydrogen atmosphere at atmospheric pressure. The hydration time ranged from several minutes to 8 h. The ultimate product (brittle, black) was obtained by keeping the scandium for 4 h in hydrogen at atmospheric pressure, followed by 4 hours at a hydrogen pressure ~ 1.5 atm.

We measured the specific magnetic susceptibility of the obtained samples and investigated the temperature dependence of χ from the temperature of liquid helium to room temperature in fields up to 10 kOe (see Fig. 1). When scandium is hydrated, the

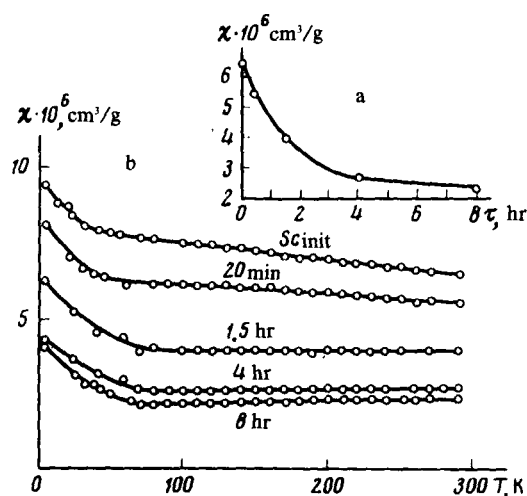


FIG. 1. a) Dependence of the magnetic susceptibility of scandium hydride on the hydration time at 600 °C. b) Temperature dependences of the susceptibility of scandium hydrides.

susceptibility decreases sharply in the samples obtained during the first 4 h, after which the changes in the susceptibility are negligible. Below 50 K, all hydrides, including scandium itself, reveal a sharp increase in the susceptibility with decreasing temperature as compared with the behavior of $\chi(T)$ in the remaining temperature region. This behavior is typical of metals with traces of ferromagnetic impurities. In the region of higher temperature (above 50 K), an intrinsic $\chi(T)$ dependence, peculiar to the metal or to the compound, appears. Substantial changes take place in the temperature coefficient of the susceptibility ($d\chi/dT$) above 50 K, namely, $d\chi/dT$ first decreases, becomes practically zero after 1.5 h of hydration, and then reverses sign and increases slightly.

It is known that the experimentally obtained susceptibility is the sum of a number of components: the spin and orbital paramagnetic susceptibilities of the conduction electrons, and the diamagnetic susceptibility of the ionic core. However, some distinguishing features of the magnetic susceptibility (the specific susceptibility and the sign of the temperature coefficient of χ) of pure metals and alloys can be qualitatively attributed to singularities of the spin component of the susceptibility, which are determined by the form of the state-density curve.

In pure scandium, the FB is located near the maximum of the state-density curve, and accordingly the magnetic susceptibility is high and $d\chi/dT < 0$ (in titanium, in which the FB is at the minimum of $N(E)$, we have $\chi = 3.23 \times 10^{-6}$ cm³/g and $d\chi/dT > 0$).

When scandium is hydrated the susceptibility decreases because the hydrogen atoms give up their electron to the conduction band of the metal, the total number of electrons is increased, and the FB shifts to the right, towards lower state densities. Therefore in pure scandium the FB should be located not simply at the maximum of the $N(E)$ curve, but on its right-hand slope. This explanation agrees with the conclusions^[4] drawn from investigations of the NMR of scandium hydrides. This is probably because of the abrupt change in the susceptibility during the initial stages of the hydration (up to 4 h). The further weak decrease of the susceptibility is connected with the increase of the content of the second phase—scandium dihydride.

In the single-phase region (hydration up to 4 h, i.e., up to 30 at.% H) the lowering of the temperature coefficient of the susceptibility can be attributed to the decrease in the slope of the $N(E)$ curve. The β phase should have a negative $d\chi/dT$, and the temperature coefficient of the second phase appears in the two-phase region (hydration 4 h and more).

Thus, our investigations of the magnetic properties of scandium hydride indicate that the hydrogen gives up its electrons to the d band of the metal. We have also shown that in the region of hcp solid solutions the magnetic properties (the value of the magnetic susceptibility and the sign of $d\chi/dT$) can be qualitatively attributed to the features of the state-density curve near the FB. These investigations confirm once more that the singularities of the spin paramagnetism, which amounts to only half of the measured magnetic susceptibility ($\chi_{sc} = 6.45 \times 10^{-6}$ cm³/g, $\chi_{sp} = 3.25 \times 10^{-6}$ cm³/g) determine the behavior of the magnetic susceptibility as the electron concentration is changed.

¹It must be stated that an investigation of the hydrides is of interest from the point of view of the mechanisms whereby superconductivity sets in. Thus, it was observed that the compound $\text{PdH}_{0.8}$ is a superconductor with rather high $T_c \sim 10 \text{ K}$.^[2]

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