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The effect of hydrogen uptake on the Hall resistivity and the electrical resistivity of gadolinium films

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Abstract

The Hall resistivity and electrical resistivity of Gd films (300 nm) coated with Pd (10 nm) are studied as a function of hydrogen pressure above and below the Gd Curie temperature. The hydrogen pressure varies from 0 to 6.0 Torr and the concentrations obtained indicate the formation of a mixed phase of Gd and GdH_2. The electrical and Hall resistivities decrease as a function of hydrogen pressure. The Hall measurements are made in magnetic fields varied from 0 to 0.60 T and they indicate a significant change on the magnetic properties of the films.

Keywords: Resistivity; Gadolinium; Gadolinium hydride; Hall resistivity

1. Introduction

Rare earths, in general, form hydrides which show drastic changes in their physical properties [1]. In the case of Gd it absorbs large amounts of hydrogen, and the formation of a trihydride has been reached. Initially, the hydrogen atoms occupy the tetrahedral sites in the hcp structure until the stoichiometric compound GdH_2 is reached. Further absorbed hydrogen atoms occupy the octahedral sites [2]. The hydrogenation changes its properties dramatically, from a magnetic metal ($T_c = 293$ K) to a semiconductor and an insulator. Also, new magnetic transitions have been observed [3]. In this paper the electrical and the Hall resistivities are reported for 300 nm thick Gd and mixed phase (Gd and GdH_2) films.

2. Experimental procedure

The films are made in a high vacuum chamber, approximately 10^{-6} Torr base pressure, by electron beam bombardment. The Gd films (99.99%) are deposited on glass substrates with proper Cu electrical contacts. Immediately a Pd (99.9%) coating is deposited on the Gd films to protect them from oxidation and to catalyze the hydrogen absorption. The films are then moved to a different chamber where the Hall and resistivity measurements are made. Hydrogen is introduced into this chamber in small steps up to approximately 6 Torr. The concentration is measured on a twin film deposited on a quartz crystal microbalance (QCM) which is placed in the same chamber. The average concentration of the Gd film with the Pd coating is calculated using an effective film mass, as explained elsewhere [4]. The hydrogen intake after each pressure increase is monitored with the QCM and the electrical resistance of the sample. When they reach stability, indicating that hydrogen uptake has ended for that particular pressure, Hall measurements are made. The Gd films thickness are of, or close to, 300 nm, while the coatings' thicknesses are of the order of 10 nm. The thicknesses are measured with the QCM with an uncertainty of 1 nm. The average resistivity of the Pd covered Gd films is 174 μΩ cm.

3. Results

The higher hydrogen concentrations obtained with the hydrogen pressures used correspond to an average H/Gd ratio of 0.8±0.1. This means that two phases, pure Gd and pure hydride GdH_2, coexist in the hydride samples. However, the strong decrease in the film's electrical resistivity, ρ, (Fig. 1) indicates that the electrical conduction is dominated by GdH_2 which has a lower resistivity, as observed by Vadja et al. [5].

The other quantity studied was the Hall resistivity, ρ_H, which is defined as E_H/J. This is a better parameter to study than the Hall coefficient when dealing with fer-

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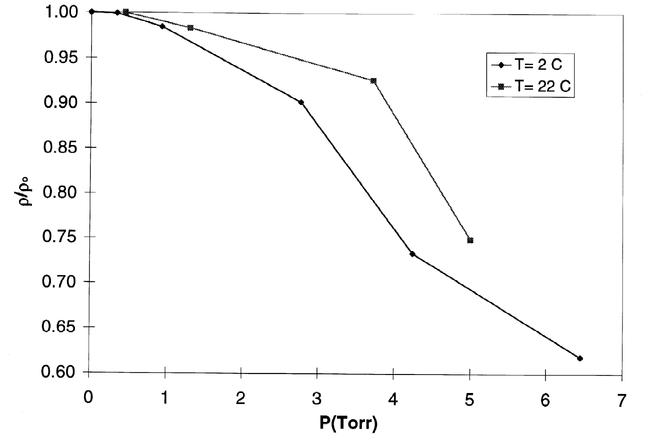

Fig. 1. Electrical resistivity as a function of hydrogen pressure above and below T_c. ρ_0 is the resistivity before hydrogenation at each temperature. The film's thickness is 296 nm for the Gd and 14 nm for the coating.

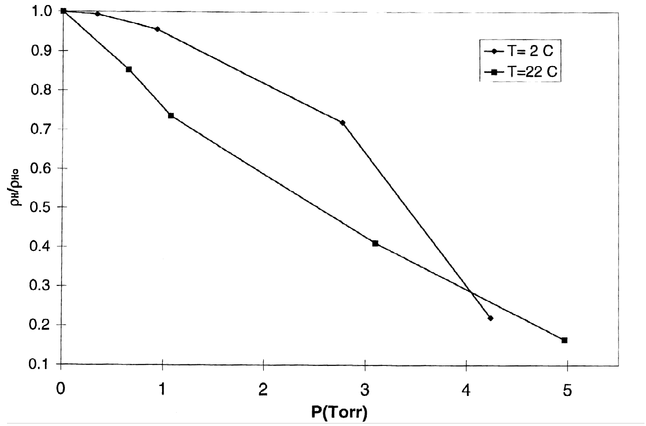

Fig. 3. Hall resistivity ratio as a function of hydrogen pressure. ρ_{H_0} is the Hall resistivity before hydrogenation at each temperature. The film's thickness is 296 nm for the Gd and 14 nm for the coating. $B=0.60$ T.

romagnetic or strong paramagnetic materials, as is the case with Gd, because it takes into account the internal field due to the magnetization [6]. The Hall resistivity can be written as $\rho_H = R_0 B + R_1 \mu_0 M$, where B is the applied magnetic field, M the magnetization, R_0 is the ordinary Hall coefficient and R_1 is the anomalous Hall coefficient, μ_0 has been included to have R_0 and R_1 in the same units. R_1 is important for ferromagnetic materials and usually it is higher than R_0 by an order of magnitude. Its origin is explained by the spin–orbit coupling of the polarized conduction electrons [6,7]. Experimentally ρ_H is calculated from $V_H t/I$, t being the film's thickness and I the electrical current. Independent measurement of R_0 and R_1 was not possible for the range of magnetic fields attainable in our laboratory.

The ρ_H curves found for pure Gd (Fig. 2) are similar to those found by other authors in this same range of magnetic fields and temperatures on bulk samples [8], indicating that these 300 nm films do not differ significantly from the bulk measurements. The larger change in ρ_H for the lower temperature is the result of a stronger magnetization below the Curie temperature, T_c, (Fig. 3). Using the bulk magnetic susceptibility, the magnetization for Gd at $T=23$ °C is 2.26×10^5 A m^{-1}, for an external field of 0.60 T. This means an equivalent extra field of 0.28 T due to the magnetization of the sample, which is not small compared with the applied field.

ρ_H in the hydrogenated samples shows a decrease down to one sixth of the value without hydrogen both above and below T_c of the Gd (Fig. 3). Also it was found that, contrary to the pure Gd case, the curve corresponding to the lower temperature shows a smaller increase (Fig. 4) as a function of magnetic field. These differences are a consequence of the changes occurring in the band structure due to the formation of the hydride, which not only affects R_0, by changing the number of conduction electrons on the Fermi level [3], but also affects R_1 by changing the spin–orbit coupling [8].

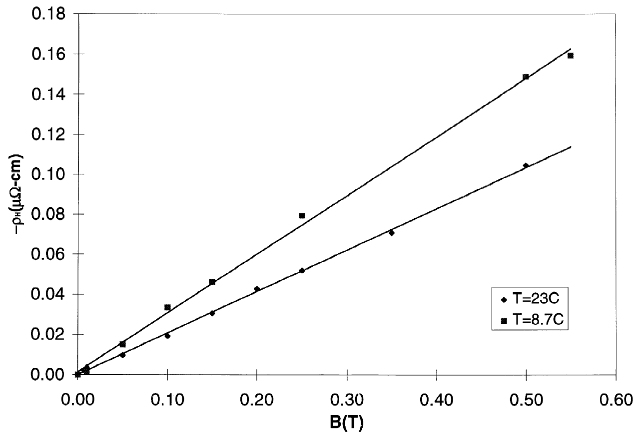

Fig. 2. Hall resistivity for the pure Gd films. Note the larger change for $T<T_c$. The Gd film thickness is 301 nm with a Pd coating of 10 nm.

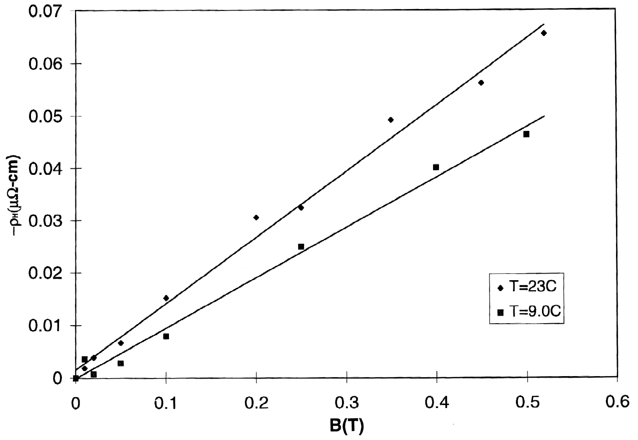

Fig. 4. Hall resistivity for hydrogenated Gd. Note the smaller scale in this figure as compared with Fig. 2. The Gd film thickness is 301 nm with a Pd coating of 10 nm. Hydrogen concentration of these films is 0.7±0.1 at.%.

4. Conclusions

As with other materials, a Pd coating allows the hydrogen to reach the Gd while it protects the Gd from oxidation. The concentration measurements and the electrical resistivity decrease indicate the formation of a hydride when the films are exposed to a hydrogen atmosphere, with the formation of a mixed phase of Gd and GdH_2. Also, ρ_H is reduced significantly with the absorption as a result of the change in the density of states at the Fermi level with the formation of the GdH_2. The ρ_H curves for the Gd films show a larger rate of change in the ferromagnetic regime ($T<T_c$) as a result of the spontaneous magnetization. The mixed phase films curves do not show the faster change at the lower temperature, indicating that the paramagnetism of the GdH_2 is dominant in the concentration studied.

Acknowledgments

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References

[1] M. Gupta and L. Schlapbach, in L. Schlapbach (ed.), *Hydrogen in Intermetallic Compounds I: Topics in Applied Physics*, Vol. 63, Springer-Verlag, Berlin, 1988, p. 139.
[2] G.G. Libowitz and A.J. Maeland, Hydrides, in K.A. Gschneidner and L. Eyring, *Handbook on the Physics and Chemistry of Rare Earths*, Vol. 3, North-Holland, Amsterdam, 1979, p. 299.
[3] P. Vadja, Hydrogenation in rare earth metals including RH_{2+x} phases, in K.A. Gschneidner and L. Eyring (eds.), *Handbook on the Physics and Chemistry of Rare Earths*, Vol. 20, Elsevier Science B.V., 1995, p. 207.
[4] D.E. Azofeifa, N. Clark, A. Amador and A. Saenz, *Thin Solid Films* (in press).
[5] P. Vadja, J.P. Burger and J.N. Daou, *J. Phys.: Condens. Matter, 3* (1991) 6267.
[6] R. Karplus and J.M. Luttinger, *Phys. Rev., 95* (1954) 1154.
[7] K.A. McEwen, Magnetic and transport properties of the rare earths, in K.A. Gschneidner and L. Eyring (eds.), *Handbook on the Physics and Chemistry of Rare Earths*, Vol. 1, North-Holland, Amsterdam, 1978, p. 411.
[8] R.S. Lee and S. Legvold, *Phys. Rev., 162* (1967) 431.