ESR OF Gd$^{3+}$ IONS IN CUBIC LAVES PHASE HYDRIDES

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Electron Spin Resonance (ESR) experiments on Gd$^{3+}$ ions in hydrides of Laves phase compounds LaRu$_2$, CeRu$_2$, ThRu$_2$, YRh$_2$ and YNi$_2$ have been performed at low temperatures. (1.5K<T<4.2K) over a wide range of hydrogen concentration. All the hydrides studied were prepared by exposing the several host samples at hydrogen pressures 1-30 atmospheres. The following interesting features were observed: i) The superconcutivity of LaRu$_2$, CeRu$_2$ and ThRu$_2$ was partially suppressed for low and intermediate hydrogen concentrations where the $\alpha$ and $\beta$ phases of the intermetallic hydrides coexist. The superconducting behaviour was completely suppressed for high hydrogen concentrations corresponding to the saturated $\beta$ phase of these hydrides. ii) For LaRu$_2$H$_x$ two well resolved ESR lines were observed at $g(\alpha)$=1.817 and $g(\beta)$=2.08 for all hydrogen concentrations ($x=0.5, 1.0, 2.0, 3.0$ and 4.5). They were associated to Gd$^{3+}$ spectra in the $\alpha$ and $\beta$ phases, respectively. Since the Gd$^{3+}$ spectrum occurs at $g=1.993$ in insulating hosts, the observed $g$-shift in the $\alpha$ phase ($\Delta g(\alpha)=-0.176$) is large and negative and changes drastically both its magnitude and sign in the saturated $\beta$ phase ($\Delta g(\beta)=0.087$). No ESR signals at intermediate g-values were observed indicating clearly that LaRu$_2$H$_x$ as well as CeRu$_2$H$_x$ and ThRu$_2$H$_x$ are inhomogeneous hydrides characterized by the existence of two phases, the $\alpha$ phase for low hydrogen concentration and the $\beta$ phase for large hydrogen concentration. This agrees with the thermodynamic results of Jacob et al. (1) for LaRu$_2$H$_x$ hydrides. iii) A dramatic reduction of the linewidth thermal broadening (Korringa rate) was observed in LaRu$_2$H$_x$ ($\Delta H_x/T=1$G/T) with respect to $\Delta H_x/T=19$G/T measured in the unhydrided LaRu$_2$. The features observed in the ESR of Gd in LaRu$_2$H$_x$ were not clearly seen in CeRu$_2$H$_x$ and ThRu$_2$H$_x$ because of the superposition of the two Gd signals in these hydrides. iv) Neither significant g-shift changes nor Korringa rate reductions were observed in the Gd ESR spectra in YRh$_2$H$_x$ and YNi$_2$H$_x$ relative to corresponding pure compounds. $\Delta g=0.002$ and $\Delta H_x/T=(2.0 \pm 0.5)$G/T for Gd$^{3+}$ in YRh$_2$H$_x$; $\Delta g=0.004$ and $\Delta H_x/T=(1.5 \pm 0.5)$ G/T in YNi$_2$H$_x$ ($x=0,1,2,3,4$ and 5). The most remarkable feature of our results is the drastic change of the Gd$^{3+}$ ESR parameters in the Ru intermetallics observed when they are hydrogenated and the small effect in the case of Rh and Ni compounds upon hydrogen take up. The Ru intermetallics are known to be strongly d-band compounds i.e. high density of electronic d-states at the Fermi level N$^d(F)$. This explains the large and negative g-shift of Gd$^{3+}$ observed in these compounds as a result of the appreciable exchange coupling of the Gd localized moment to the d-band electronic susceptibility of the host. The sign reversal of the g-shift in the Ru intermetallic hydrides suggest that the Ru d-bands are filled by electrons transferred from the hydrogen atoms as predicted by the protonic hydrogen model. This interpretation is supported by susceptibility data which indicate a marked reduction in the paramagnetic susceptibility of LaRu$_2$ when it is hydrided to LaRu$_2$H$_x$. Finally, the very small effect (if any) of the absorbed hydrogen on the ESR parameters of Gd$^{3+}$ ions in Rh and Ni compounds is not well understood at the present. Detailed investigations are in progress in order to improve our understanding of the ESR properties of these hydrides.

1. I. Jaboc and D. Shaltial, J.L.C. Metals, 65,117(1979); 2. G.X. Tassema, Thèse de 3 Cycle, Grenoble (1979). This work was supported by CNPq and FAPESP (Brasil)