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. ($1.5K \leq T \leq 4.2K$) 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 superconcuctivity of $LaRu_2$, $CeRu_2$ and $ThRu_2$ was partially supressed for low and intermediate hydrogen concentrations where the α and β phases of the intermetallic hydrides coexist. The superconducting behaviour was completely suppressed for high hydrogen concentrations corresponding to the saturated β phase of these hydrides. ii) For $LaRu_2H_x$ 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 α and β phases, respectively. Since the Gd^{3+} spectrum occurs at $g = 1.993$ in insulating hosts, the observed g-shift in the α phase ($\Delta g(\alpha)=-0.176$) is large and negative and changes drastically both its magnitude and sign in the saturated β phase ($\Delta g(\beta)=0.087$). No ESR signals at intermediated g-values were observed indicating clearly that $LaRu_2H_x$ as well as $CeRu_2H_x$ and $ThRu_2H_x$ are inhomogeneous hydrides characterized by the existence of two phases, the α phase for low hydrogen concentration and the β phase for large hydrogen concentration. This agrees with the thermodynamic results of Jacob et al.[1] for $LaRu_2H_x$ hydrides. iii) A dramatic reduction of the line width thermal broadening (Korringa rate) was observed in $LaRu_2H_x$ ($\Delta H_k/T \simeq 1G/T$) with respect to $\Delta H_k/T \simeq 19G/T$ measured in the unhydrided $LaRu_2$. The features observed in the ESR of Gd in $LaRu_2H_x$ were not clearly seen in $CeRu_2H_x$ and $ThRu_2H_x$ 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_2H_x and YNi_2H_x relative to corresponding pure compounds. $\Delta g=0.002$ and $\Delta H_k/T=(2.0\pm0.5)G/T$ for Gd^{3+} in YRh_2H_x; $\Delta g=0.004$ and $\Delta H_k/T \simeq (1.5\pm0.5)$ G/T in YNi_2H_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 intermetallic are known to be strongly d-band compound i.e.high density of electronic d-states at the Fermi level $N^d(E_F)$. This explain 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 reverse of the g-shift in the Ru intermetallic hydrides suggest that the Ru d-bands are filled by electrons transfered 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 $LaRu_2H_x$[2]. 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 3º Cycle, Grenoble (1979). This work was supported by CNPq and FAPESP (Brasil)