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Observation of the $\text{Er}^{3+} \Gamma_8$ groundstate ESR-resonance in YAl$_2$ single crystals (*)

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Résumé. — On montre que le niveau fondamental de $\text{Er}^{3+}$ dans YAl$_2$ est un niveau $\Gamma_8$. Les expériences ont été menées pour une concentration d’environ 1 000 ppm. On obtient le paramètre de champ cristallin, $x$, égal à $+0.23$.

Abstract. — It is shown that the groundstate of $\text{Er}^{3+}$ in YAl$_2$ is a $\Gamma_8$. The experiments were performed for a concentration of approx. 1 000 ppm. The crystal field parameter yields $x = +0.23$.

The cubic Laves phases $X_{1-x}\text{Er}_x\text{Al}_2$ ($X = \text{Y, La}$) have been the subject of many investigations for the determination of the crystal field (CF) splitting of $\text{Er}^{3+}$. Samples with different concentrations, $x$, have been investigated by susceptibility and specific heat measurements as well as inelastic neutron scattering [1] and electron spin resonance (ESR) [2]. The experimental results published up to now do not show a consistent levelscheme — or in the notation of LLW [3], the $x$ and $W$ values are completely inconsistent. The ESR measures magnetic dipole transitions within the groundstate manifold. This determines the groundstate properties very precisely. $\Gamma_0$, $\Gamma_2$ or the anisotropic $\Gamma_8$ resonance can be distinguished easily. For the latter, single crystals are needed, because of the anisotropy of the resonance spectrum [4, 5]. Most of the experimental technique mentioned above are limited to a concentration of magnetic impurities down to $10^{-6}$ to $10^{-5}$. At these concentrations interaction effects are still present and may mask the pure CF.

Here we report ESR results on samples doped with $x = 1.000$ and $1.500$ ppm. This low concentration justifies the assumption of non interacting ions. The single crystals were grown from the melt. The experiments were performed at 10 and 35 GHz. Figure 1 shows two resonance transitions: the field for resonance, linewidth and intensity depending on the orientation. At $g = 2$ a background signal is produced by the empty cavity. The dashed lines are single line fits using a Dysonian lineshape. At 10 GHz similar signals were recorded, but because of some crystallographic imperfections for larger size crystals, the resonance signal became broader and the signal to noise ratio was worse.

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Fig. 1. — ESR-Signal for Er concentration at 1 500 ppm. $\theta$ being the angle between the applied field and the cubic axis.

Figure 2 shows the possible level scheme for $J = 15/2$ and cubic symmetry. From our results in figure 1 there are only two solutions: $\Gamma_8^{(1)} W > 0$ and $0.6 \leq x \leq 1$ or $\Gamma_8^{(3)} W < 0$ and $-1 \leq x \leq 0.8$. Furthermore we can exclude low lying (few deg. K) excited levels. In such a case the Zeeman interaction would not be linear in the frequency because of an
Taking the conduction electrons into consideration the complete Hamiltonian is given by [6]:

$$\mathcal{H} = W \left[ x \frac{O_x(J)}{F_z} + (1 - |x|) \frac{O_y(J)}{F_z} \right] + g_J \mu_B \mathbf{H} \cdot \mathbf{J} + J_{\text{eff}} (g_J - 1) \mathbf{J} \sigma.$$

Here we assume an isotropic exchange interaction $J_{\text{eff}}$. It acts on the magnetic impurities as an effective field and can be described within the groundstate manifold as an effective change of the $g$-value. The fit in figure 3 yields $g_{\text{exp}}/g_{\text{theor}} = 1.0868$

$$g_{\exp}/g_{\text{theor}} = 1 + \frac{g_J - 1}{g_J} \cdot N(E_F) J_{\text{eff}}.$$

This means a positive $g$-shift of $N(E_F) J_{\text{eff}} = 0.5$. The value is quite large compared to Gd : YAl$_2$ $N(E_F) J_{\text{eff}} = 0.07$ [7]. In our analysis we have assumed an isotropic exchange. The difference at a factor 7 for Er and Gd may indicate that an anisotropic exchange analysis is needed. Pellisson [8] has shown that this is needed for Er doped into Pt.

Our results are in rough agreement with recent inelastic neutron scattering data on an 8% Er sample [10], the experiments yield $x = +0.36$ and $W' = -0.046$ meV. The difference in the two $x$-values is out of the experimental error bars and can have several reasons: 1) there exists a real concentration dependence of the CF parameters, given by the change of the lattice constant and/or the change of the effective screening charge by substituting a RE ion on an Y-site. 2) For concentration of several % RE ions an internal field distribution surely is produced as well as an RKKY coupling of the RE ions will be present. (In earlier neutron scattering work [9] a quasielastic peak was detected.) This will affect the $\Gamma_8$ ground state and the analysis.

We have shown that ESR is a convenient technique to determine CF ground states at very low impurity concentrations. This in accord with neutron scattering for moderate RE concentrations yields a unique set of CF parameters. Susceptibility and other bulk measurements are at least in this system very insensitive (see figure 5 in Ref. [9]) to different levels schemes. Further experiments to determine the thermal broadening, a possible concentration dependent $x$-value and the system Er : LaAl$_2$ are under current investigation.

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References


