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EFFECTS OF THE EXCITED CRYSTALLINE FIELD STATES IN THE *ESR* OF *Er:Au*

BY

K. BABERSCHKE

Fachbereich Physik, Freie Universität Berlin, Germany

INTRODUCTION

The effect of excited crystalline field states on the relaxation rate of the ground state is well known in insulators. This "Orbach process" is strongly temperature dependent being determined by the energy splitting Δ between the ground state and the first excited crystalline field (c.f.) state [1].

In 1969 Williams and Hirst [2] have determined the c.f. splitting of several rare earth ions in *Au* and *Ag*. In addition in 1969 Hirst [3] has given the general formula for the relaxation rate in a multilevel system of a localized moment, which is coupled to the conduction electrons. The most favoured system for detecting this effect by *ESR* will be *Er:Au*. Up to now only the resonance of *Er*, *Dy*, *Yb* (besides the *S*-state ions) was detected in metals. In most of the host metals *Er* shows the smallest thermal broadening. The smallest residual width was observed in *Er:Au* [4]. Both facts favour this system because one is able to follow the resonance signal up to high temperatures.

EXPERIMENTAL RESULTS

Previous *ESR* measurements [4] were performed at temperatures below 4.2° *K*. In this temperature range the thermal broadening is $b = 2.7 \text{ G/K}$. The g -value of $g = 6.8$ indicates a Γ_7 ground state, which is in agreement with the static susceptibility measurements [2]. Figure 1 shows the resonance signal at 4 and 14° *K*. At 4° *K* one observes the hyperfine splitting. At higher temperatures this is smeared out and the error on determining the linewidth increases. Depending on the *Er* concentration we were able to follow the resonance signal up to 20-30 *K*. Figure 2 shows a break at roughly 6 *K* in the linear temperature dependence of the linewidth. This is attributed to an additional relaxation rate by excited c.f. states. Measurements on other systems *Er*, *Dy* in *Ag*, *Ir*, *Th* are performed and will be published elsewhere [5, 6].

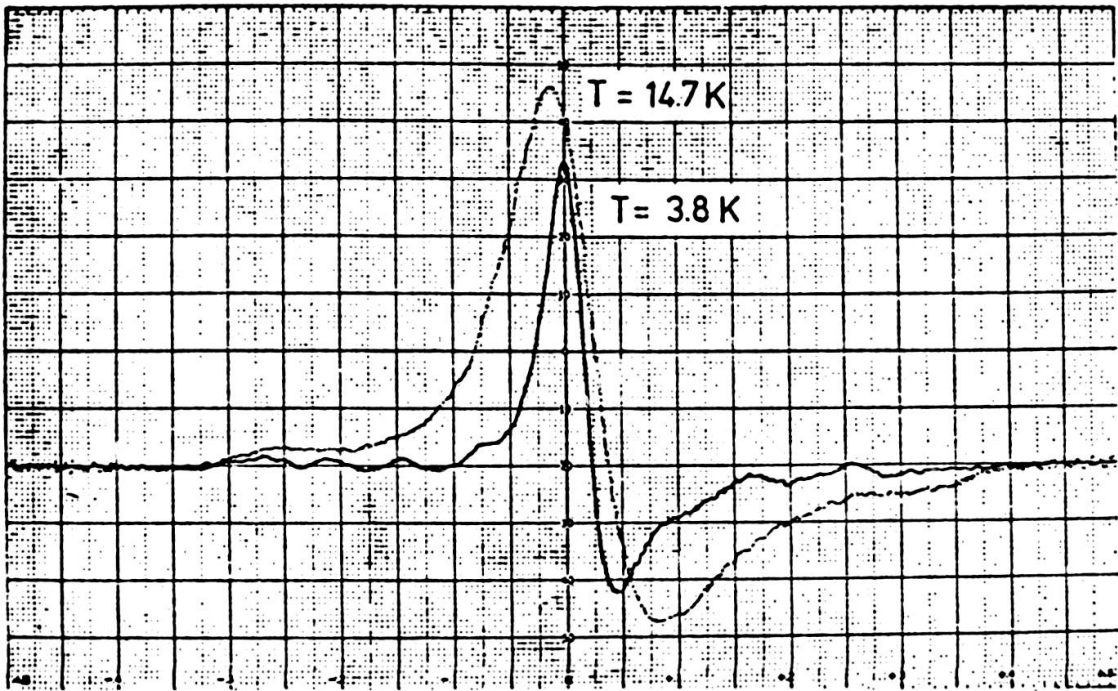


FIG. 1. — ESR signal of 1000 ppm *Er:Au* at X-band and 100 KHz modulation frequency. $H_0 = 984$ G, $H_{sweep} = 1$ kG. The spectra at 14.7°K is amplified by a factor of 5.

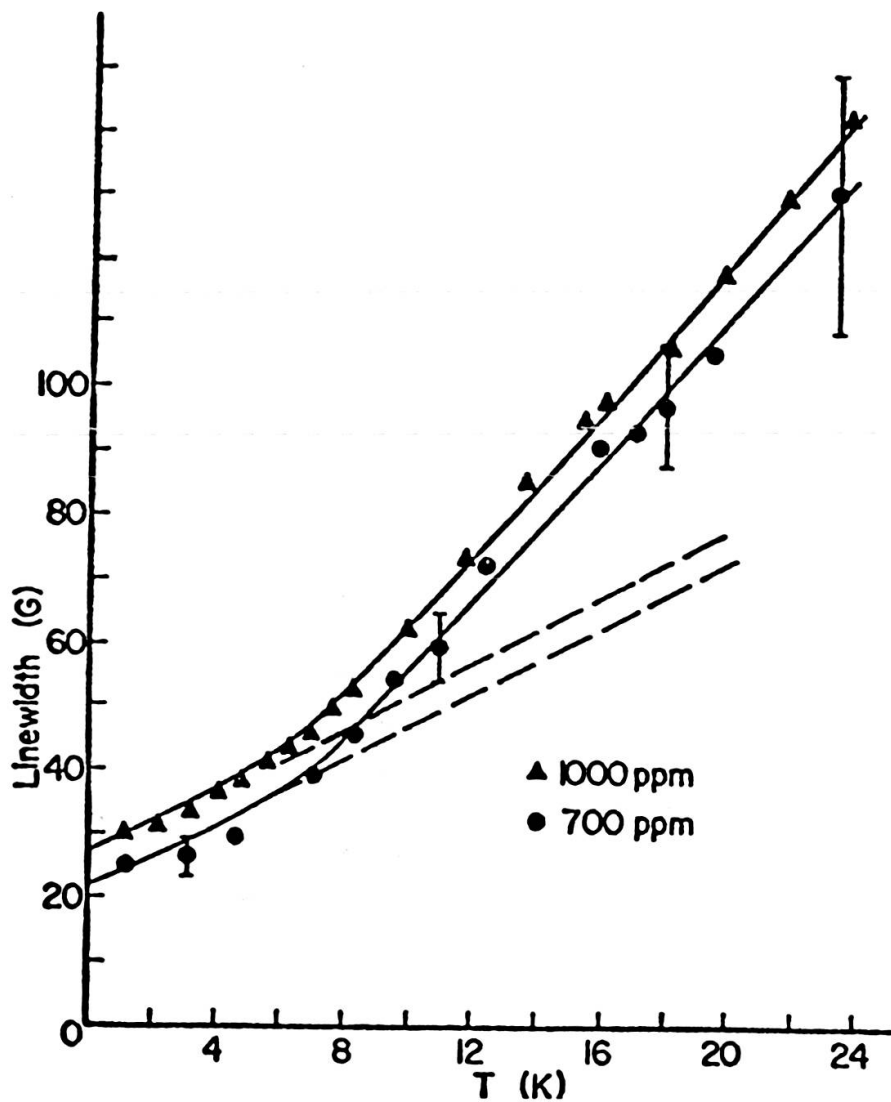


FIG. 2. — Linewidth as a function of temperature. Different *Er* concentrations show different residual linewidth but the same slope.

DISCUSSION

Susceptibility measurements show the first excited level to be Γ_8 . Labelling the ground state by $|\pm\rangle$ and the excited Γ_8 by $|i\rangle$, we find the thermal broadening ΔH .

$$\Delta H = \frac{\pi}{2g_{\text{eff}} \mu_B} [(g_J - 1) \eta(E_F)]^2 \langle J^2(q) \rangle (A + B + C) \quad (1)$$

$$A = (\langle + | J_z | + \rangle - \langle - | J_z | - \rangle)^2 k_B T,$$

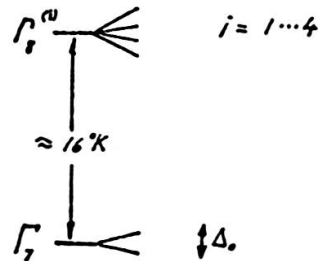
$$B = \frac{1}{2} |\langle + | J_+ | - \rangle|^2 \{f(\Delta_0) + f(-\Delta_0)\}$$

$$C = \frac{1}{2} \sum_i \left\{ (|\langle + | J_+ | i \rangle|^2 + |\langle + | J_- | i \rangle|^2) f(\Delta_i^+) + (|\langle - | J_+ | i \rangle|^2 + |\langle - | J_- | i \rangle|^2) f(\Delta_i^-) + 2|\langle + | J_z | i \rangle|^2 f(\Delta_i^+) + 2|\langle - | J_z | i \rangle|^2 f(\Delta_i^-) \right\}$$

$$\Delta_0 = E(\Gamma_7, +) - E(\Gamma_7, -);$$

$$\Delta_i^\pm = E(\Gamma_8, i) - E(\Gamma_7, \pm);$$

$$f(\Delta) = \frac{\Delta}{e^{\Delta/k_B T} - 1}$$



There are 3 contributions A , B , C to ΔH . A and B are caused only by the Γ_7 ground state. “ A ” is the so-called “frequency modulation” part of the relaxation rate [7], which becomes exactly zero at zero temperature. The contribution “ B ” becomes constant at zero temperature and proportional to $\Delta_0 = g\beta_B H_0$. The contribution of the excited c.f. state is given by the term “ C ”. Assuming an overall splitting of $105^\circ K$ [2] we were able to fit the experimental data to the theoretical formula with $\Delta = 16 \pm 6^\circ K$. This is in good agreement with the splitting determined by susceptibility measurements [2]. The big errorbar is caused by the unsensitivity of the relaxation rate on Δ as shown in Figure 3. Similar effects on the relaxation rate in $Er: Au$ are measured by Mössbauer technique [9].

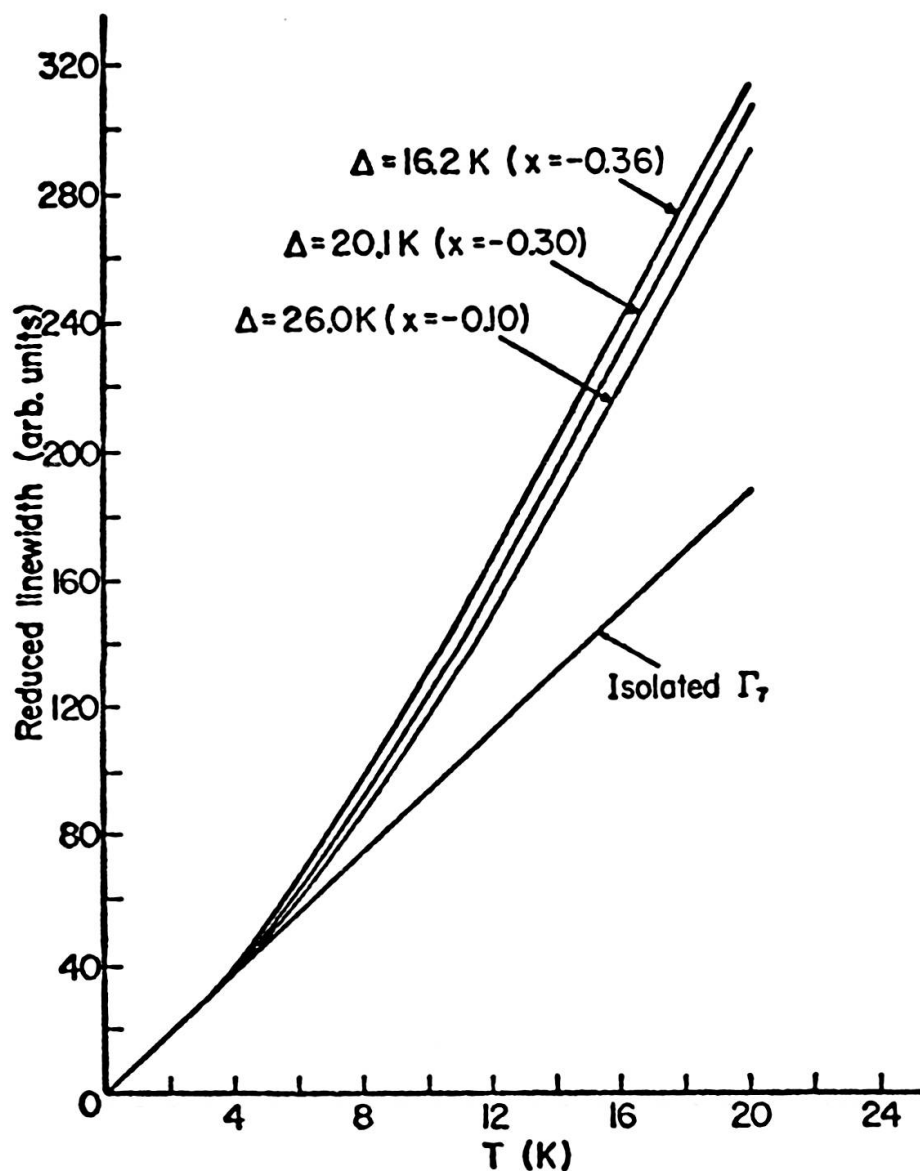


FIG. 3. — Thermal broadening calculated by formula (1). x is a measure of the ratio of the fourth- and sixth-order c.f. parameter [8].

SUMMARY

We reported for the first time the effect of excited c.f. states on the relaxation rates of localized moments in metals by *ESR*. The results show that the c.f. in metals acts in a similar way than in insulators. Furthermore the perfect agreement between theory and data shows by itself that the *ESR* signal is a bulk measurement of the *Er:Au* alloy which means surface effects are small. Further results on the relaxation rate and on the g -factor by excited c.f. states are to be published elsewhere [5, 6].

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