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ESR AND THE MAGNETIC PROPERTIES OF THE $GdAl_2$-$GdCo_2$ SYSTEM*

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The X-ray, magnetic and ESR study of ferromagnetic $Gd(Co_xAl_{1-x})_2$ type compounds were performed for the paramagnetic region at 77–400 K. The transition temperatures, effective moments, thermal broadening of the resonance line $\partial \Delta H/\partial T$, and g factor were measured. It was found that $\partial \Delta H/\partial T$ is independent of the $GdCo_2$ concentration in $GdAl_2$ for $GdCo_2$ contents exceeding 10%. For Co containing specimens the g factor for Gd^{3+} was decreased compared to that for $GdAl_2$.

The structural and magnetic properties of the $GdCo_2$-$GdAl_2$ series were studied by Oesterreicher and Wallace [1]. The $GdAl_2$ compound was studied also by the ESR method [2–6]. This paper presents the results of the magnetic and ESR measurements for the $Gd(Co_xAl_{1-x})_2$ compounds. The alloys investigated were prepared by arc-melting of the starting metals Gd(3N), Co(3N), Al(5N) in Ar. The X-ray study performed at room temperature using powder samples in Fe-$k\alpha$ radiation, showed that the materials were single-phase. $GdAl_2$-$GdCo_2$ crystallizes in the Laves phase $C14$ or $C15$. The Hexagonal Laves phase $C14$ was observed for 30—90% $GdCo_2$ in $GdAl_2$, for less and greater concentrations the cubic Laves phase $C15$ was noticed (Table I).

The magnetic measurements were performed by the Faraday method at 77—450 K. The magnetic phase transition temperatures were determined from the temperature dependence of magnetisation at up to 800 Oe. The Curie-Weiss constant and magnetic effective moment were determined from the reciprocal of the temperature dependence of magnetic susceptibility. It was found that the transition temperature of $GdAl_2$ was 171 K. The literature values of T_c for $GdAl_2$ differ enough from each other and cover the range 150—180 K [2–6]. The authors had decided to check the dependence of the transition temperature on the preparation of alloys, i.e. the arc temperature, sample stoichiometry, contamination of starting materials, oxygen content in the inert furnace atmosphere and soaking time. The sample prepared by these methods showed Curie temperatures

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TABLE I

mol % $GdCo_2$ in $GdAl_2$	Laves phase	T_c [K]	θ [K]	μ_{eff} [μ_B]	$\partial\Delta H/\partial T$ [Oe/deg]	g
0	*C*15	171	164	8.07	2.9±0.3	1.94±0.04
10	*C*15	153	130	7.41	7.0±0.3	1.87
20	*C*15	110	97	8.02	7.0	1.88
25	*C*15	111	83	8.00	7.0	1.87
30	*C*14	90	81	7.95	6.9	1.87
40	*C*14	109	94	7.58	6.9	1.81
50	*C*14	117	114	7.3	7.0	$\geqslant$1.8
60	*C*14	145	134	6.42		
70	*C*14	250	235	5.08		
80	*C*14	339				
90	*C*14	355	does not follow the Curie-Weiss law			
100	*C*15	400				

from 168 to 181 K. No samples with smaller transition temperatures were obtained. The presence of oxygen in the argon furnace atmosphere exerted a most significant effect on the deviation of T_c from the value of 171, by increasing this parameter. The other factors changed T_c within the ±3 K range. The temperature 171 K seems to be a real transition temperature for the single-phase ordered compound $GdAl_2$.

Fig. 1 shows the dependence of the Curie temperature and effective magnetic moments on the $GdCo_2$ concentration in the compounds studied. The Curie temperature changes from 171 K for $GdAl_2$ to 409 K for $GdCo_2$ reaching a minimum at ca. 30% $GdCo_2$. The effective moment within the 0—30% Co concentration range does not change within the error limits, except for 10% Co and attains ca. 8 μ_B. This fact is presumably connected with the attenuation of the moment on Co ions (μ_{eff} for Gd^{3+} ions in other compounds is 8 μ_B). At higher $GdCo_2$ concentrations the effective moment decreases down to 5 μ_B

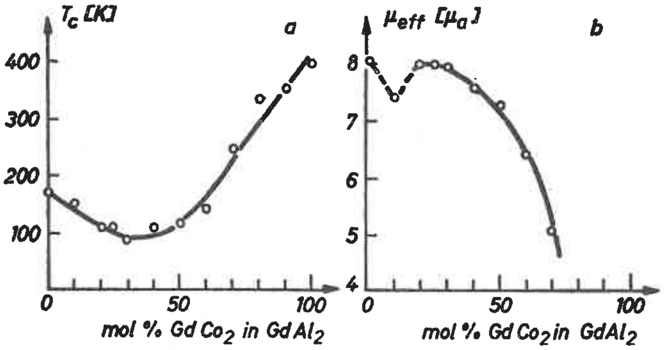

Fig. 1. a) The dependence of the Curie temperature T_c on $GdCo_2$ concentration; b) effective magnetic moments as a function of the $GdCo_2$ concentration

for the sample containing 70% Co. The sample containing greater than 75% Co did not follow the Curie-Weiss law. Since the effective moment is in fact a resultant moment for a paramagnetic molecule, it was rather an increase in μ_{eff} with Co ion concentration that was to be expected (the effective moment for Co ions in other compounds is 3.14 μ_B [9]). This can be attributed to the presence of a phase transition at higher temperatures (above 450 K), due to which a sample cannot be in a paramagnetic state within the temperature range studied. A diminution of the effective moment can also be due to a decrease in the magnetic moment on Gd, which can be connected with a change in the number of 4*f* electrons. The values of T_c and μ_{eff} are given in Table I. These results, however, are not in conformity with those previously obtained [1].

The ESR measurements were performed within the X-band at 90—300 K. The observed resonance lines for Gd^{3+} ions were asymmetrical. Their analysis was performed

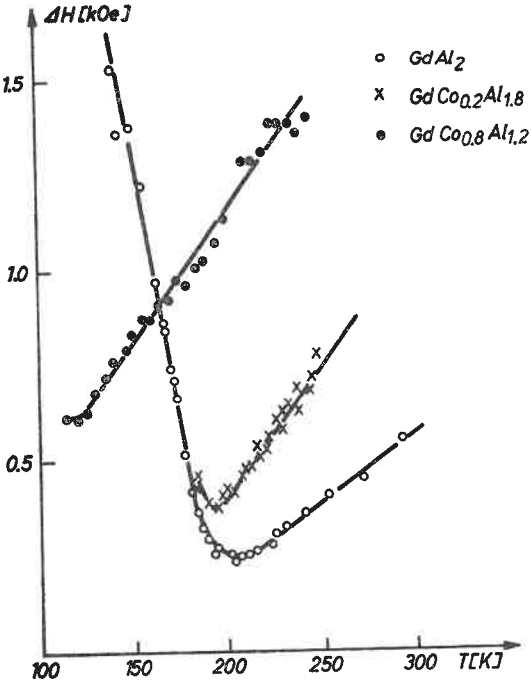

Fig. 2. The temperature dependence of the resonance linewidth for different concentrations of Co in $Gd(Co_xAl_{1-x})_2$

by the method described by Peter et al. [2]. The width of the ESR line decreased linearly with temperature attaining a minimum (several hundred Oe), but near the transition temperature a rapid increase in width, characteristic of the paramagnetic-ferromagnetic transition, was noticed. In metallic compounds containing magnetic ions with spin $\boldsymbol{S}$ and conduction electrons with spin $\boldsymbol{s}$, there occurs an exchange interaction between these spins and the hamiltonian:

$$\mathcal{H} = -J\boldsymbol{S}\cdot\boldsymbol{s}$$

where J is exchange integral. This interaction accounts for the shifting of the g factor and thermal broadening of the ESR line. The ESR results obtained for Gd^{3+} ions in $GdAl_2$ agree with earlier works [2, 6], i.e. the thermal broadening is 2.9 Oe/deg and the g factor is 1.94 ± 0.04. Hence the g shift is negative ($g_{\mathrm{dielectric}} = 1.998$), which, according to the above formula, gives negative J values. Substituting other paramagnetic ions e.g. Co in place of nonmagnetic Al ions, one can expect a change in the exchange integral and hence a change in the shift Δg and $\partial \Delta H / \partial T$. Similar changes were observed for the compounds investigated. A few examples of the $\Delta H(T)$ dependences are given in Fig. 2. The thermal broadening of the resonance line for all the Co-containing alloys studied approximately equals 7 Oe/deg which is significantly less than the Korringa width which equals 60 Oe/deg for Gd^{3+} in $LaAl_2$ [8]. The addition of Co to $GdAl_2$ caused a diminution of the g factor, which was to be expected. A study of alloys containing up to 10% $GdCo_2$ in $GdAl_2$, near the structural transition points, and exact calculation of J will make possible a more precise interpretation of interactions in $Gd(Co_xAl_{1-x})_2$ type compounds.

REFERENCES

[1] H. Oesterreicher, W. E. Wallace, *J. Less Common Metals* **1**, 91 (1967).

[2] M. Peter, D. Shaltied, J. H. Wernick, J. B. Mock, R. C. Scherwood, *Phys. Rev.* **126**, 1395 (1962).

[3] M. P. Kawatra, J. A. Mydosh, J. I. Budnick, *Phys. Lett.* **24A**, 421 (1967).

[4] H. J. Van Dall, K. H. J. Buschow, *Solid State Commun.* **7**, 217 (1969).

[5] A. Hacker, R. Gupta, M. L. Sheppard, *Phys. Status Solidi* **9**, 601 (1972).

[6] R. H. Taylor, B. R. Coles, *J. Phys. F* **5**, 121 (1975).

[7] I. Ursu, E. Burzo, *J. Magn. Resonance* **8**, 274 (1972); D. Davidov, R. Orbach, C. Rettori, D. Shaltied, L. J. Tao, B. Richs, *Phys. Lett.* **A35**, 339 (1971); J. Korringa, *Physica* **16**, 601 (1950); T. Moriya, *J. Phys. Soc. Jap.* **18**, 516 (1963).

[8] D. Davidov, A. Chełkowski, C. Rettori, A. Orbach, *Phys. Rev.* **B7**, 1029 (1973); R. H. Taylor, *J. Phys. F* **3**, L 110 (1973); G. Koopman, V. Engel, K. Baberschke, S. Huffner, *Solid State Commun.* **11**, 1197 (1972).

[9] R. S. Tebble, D. J. Craik, *Magnetic Materials*, John Wiley and Sons LTD., London 1969.